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(54) **Dielectric ceramic composition and monolithic ceramic capacitor using same**

(57) The present invention provides a dielectric ceramic composition containing 100 parts by weight of essential component represented by  $(\text{BaO})_m\text{TiO}_2 + \text{M}_2\text{O}_3 + \text{R}_2\text{O}_3 + \text{BaZrO}_3 + \text{MgO} + \text{MnO}$  (wherein  $\text{M}_2\text{O}_3$  represents  $\text{Sc}_2\text{O}_3$  and/or at least one of  $\text{Eu}_2\text{O}_3$ ,  $\text{Gd}_2\text{O}_3$ ,  $\text{Tb}_2\text{O}_3$  and  $\text{Dy}_2\text{O}_3$ ) and 0.2 to 3.0 parts by weight of the side components represented by  $\text{Li}_2\text{O} \cdot (\text{Si}, \text{Ti})\text{O}_2 \cdot \text{MO}$  (wherein MO represents  $\text{Al}_2\text{O}_3$  and/or  $\text{ZrO}_2$ ) or  $\text{SiO}_2 \cdot \text{TiO}_2 \cdot \text{XO}$  (wherein XO represents at least one of  $\text{BaO}$ ,  $\text{CaO}$ ,  $\text{SrO}$ ,  $\text{MgO}$ ,  $\text{ZnO}$  and  $\text{MnO}$ ), and a ceramic capacitor using the same.

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## Description

## BACKGROUND OF THE INVENTION

## 1. Field of the Invention

The present invention relates to a dielectric ceramic composition and monolithic ceramic capacitor using the same.

## 2. Description of the Related Art

The conventional ceramic capacitor is usually produced by the following process.

First, a sheet of a dielectric material coated with an electrode material to serve as inner electrodes on its surface is prepared. A material essentially composed of  $\text{BaTiO}_3$  is used for the dielectric material. Then, the sheet of the dielectric material coated with this electrode material is laminated with heat-pressing to a monolithic body followed by firing at 1250 to 1350 °C in an atmospheric environment to obtain a ceramic monolithic body having inner electrodes. A monolithic ceramic capacitor is obtained by glazing outer electrodes electrically connected to inner electrodes.

Noble metals such as platinum, gold, palladium or silver have been conventionally used for the material of the inner electrode of this monolithic ceramic capacitor. However, these electrode materials are expensive while having excellent characteristics, rendering the production cost to be increased. Therefore, a monolithic capacitor using base metals such as Ni in the inner electrode is currently proposed to reduce the production cost, its application in the market being steadily increasing.

## SUMMARY OF THE INVENTION

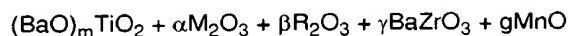
In the trend to make electronic appliances compact, high performance and low price, the monolithic capacitor is strongly required to be low price, to be improved in insulation durability, insulating property and reliability, and to have a large capacitance. Although it is advantageous for reducing the price of the electronic appliances to use an inexpensive monolithic ceramic capacitor in which nickel is used for the inner electrode, a problem that the insulation resistance, insulation durability and reliability are extremely deteriorated when the electronic appliances are used under a high electric field strength because conventional dielectric ceramic materials are designed on the premise that they are used under a low electric field strength. In other words, there has been no monolithic ceramic capacitor being able to use under a high electric field strength along with using nickel for the inner electrode.

For example, while the dielectric materials disclosed in Japanese Examined Patent Publication No. 57-42588 and Japanese Unexamined Patent Publication No. 61-101459 can display a large dielectric constant, the grain size of the dielectric ceramic is large, thereby exhibiting deficiencies such that the insulation durability of the monolithic ceramic capacitor becomes low when it is used under a high electric field strength or the mean life span under the high temperature load test becomes short.

In the dielectric material disclosed in Japanese Examined Patent Publication No. 61-14611, there was a deficiency that the dielectric constant, or the electrostatic capacitance, is extremely lowered when the capacitor is used under a high electric field strength, although its dielectric constant obtained under a low electric field strength is as high as 2000 to 2800. It was also a deficiency that the insulation resistance is low.

The object of the present invention is to provide a dielectric ceramic composition capable of forming, for example, dielectric ceramic layers of a monolithic ceramic capacitor, wherein the insulation resistance represented by a product with the electrostatic capacitance (a product CR) is as high as 4900 to 5000  $\Omega \cdot \text{F}$  or more and 200  $\Omega \cdot \text{F}$  or more at room temperature and 150 °C, respectively, when the capacitor is used under a high electric field strength of, for example, as high as about 10 kV/mm, along with having a small voltage dependence of the insulation resistance, being excellent in stability of the electrostatic capacitance against DC bias voltage, being high in the insulation durability besides the temperature characteristics of the electrostatic capacitance satisfying both of B-level characteristic standard stipulated in the JIS Standard and X7R-level characteristic standard stipulated in the EIA standard and being excellent in weather resistance performance shown by a high temperature load test and high humidity load test. Another object of the present invention is to provide a monolithic ceramic capacitor whose inner electrode is constructed of Ni or Ni alloys along with using such dielectric ceramic composition as a dielectric ceramic layer.

In a first aspect, the present invention provides a dielectric ceramic composition comprising barium titanate containing 0.02% by weight or less of alkali metal oxides, at least one of either scandium oxide or yttrium oxide, at least one kind of compound selected from europium oxide, gadolinium oxide, terbium oxide and dysprosium oxide, and barium zirconate and manganese oxide, and containing an essential component represented by the following composition formula;

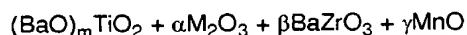


(wherein  $\text{M}_2\text{O}_3$  represents at least one of either  $\text{Sc}_2\text{O}_3$  or  $\text{Y}_2\text{O}_3$  and  $\text{R}_2\text{O}_3$  represents at least one of the compound selected from  $\text{Eu}_2\text{O}_3$ ,  $\text{Gd}_2\text{O}_3$ ,  $\text{Tb}_2\text{O}_3$  and  $\text{Dy}_2\text{O}_3$ ,  $\alpha$ ,  $\beta$ ,  $\gamma$  and  $g$  representing mole ratio in the range of  $0.001 \leq \alpha \leq 0.05$ ,  $0.001 \leq \beta \leq 0.05$ ,  $0.005 \leq \gamma \leq 0.06$ ,  $0.001 < g \leq 0.13$  and  $\alpha + \beta \leq 0.06$  with  $1.000 < m \leq 1.035$ ),

along with containing 0.2 to 3.0 parts by weight of either the first or second side component relative to 100 parts by weight of the essential component, wherein the first side component is an oxide represented by  $\text{Li}_2\text{O} - (\text{Si}, \text{Ti})\text{O}_2 - \text{MO}$  (wherein MO is at least one of  $\text{Al}_2\text{O}_3$  or  $\text{ZrO}_2$ ) and the second side component is an oxide represented by  $\text{SiO}_2 - \text{TiO}_2 - \text{XO}$  (wherein XO is at least one of the compound selected from BaO, CaO, SrO, MgO, ZnO and MnO).

In the dielectric ceramic composition described above, the essential component may further contain  $h$  mole ratio of magnesium oxide, where  $0.001 < g \leq 0.12$ ,  $0.001 < h \leq 0.12$  and  $g + h \leq 0.13$

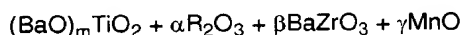
In the dielectric ceramic composition according to another aspect of the present invention, the essential component may be represented by the following composition formula;



(wherein  $\text{M}_2\text{O}_3$  represents at least one of either  $\text{Sc}_2\text{O}_3$  or  $\text{Y}_2\text{O}_3$ , where  $\alpha$ ,  $\beta$  and  $\gamma$  representing mole ratio in the range of  $0.001 \leq \alpha \leq 0.06$ ,  $0.005 \leq \beta \leq 0.06$  and  $0.001 < \gamma \leq 0.13$  with  $1.000 < m \leq 1.035$ ).

The essential component may further contain  $g$  mole ratio of magnesium oxide, where  $0.001 < \gamma \leq 0.12$ ,  $0.001 < g \leq 0.12$  and  $\gamma + g \leq 0.13$

According to a different aspect of the present invention, the essential component may be represented by the following composition formula;



(wherein  $\text{R}_2\text{O}_3$  represents at least one kind of compound selected from  $\text{Eu}_2\text{O}_3$ ,  $\text{Gd}_2\text{O}_3$ ,  $\text{Tb}_2\text{O}_3$ ,  $\text{Dy}_2\text{O}_3$ ,  $\text{Ho}_2\text{O}_3$ ,  $\text{Er}_2\text{O}_3$ ,  $\text{Tm}_2\text{O}_3$  and  $\text{Yb}_2\text{O}_3$ , where  $\alpha$ ,  $\beta$  and  $\gamma$  representing mole ratio in the range of  $0.001 \leq \alpha \leq 0.06$ ,  $0.005 \leq \beta \leq 0.06$  and  $0.001 < \gamma \leq 0.13$  with  $1.000 < m \leq 1.025$ ).

The essential component may further contain  $g$  mole ratio of magnesium oxide, where  $0.001 \leq \beta \leq 0.06$ ,  $0.001 < \gamma \leq 0.12$ ,  $0.001 < g \leq 0.12$  and  $\gamma + g \leq 0.13$ .

In the dielectric ceramic composition described above, it is preferable that the first side component, when its composition is represented by  $x\text{Li}_2\text{O} - y(\text{Si}_w\text{Ti}_{1-w})\text{O}_2 - z\text{MO}$  (wherein  $x$ ,  $y$  and  $z$  represent mol% and  $w$  is in the range of  $0.30 \leq w \leq 1.00$ ), falls within or on the boundary lines of the area surrounded by straight lines connecting each point indicated by A ( $x = 20$ ,  $y = 80$ ,  $z = 0$ ), B ( $x = 10$ ,  $y = 80$ ,  $z = 10$ ), C ( $x = 10$ ,  $y = 70$ ,  $z = 20$ ), D ( $x = 35$ ,  $y = 45$ ,  $z = 20$ ), E ( $x = 45$ ,  $y = 45$ ,  $z = 10$ ) and F ( $x = 45$ ,  $y = 55$ ,  $z = 0$ )

(when the composition falls on the straight line of A - F,  $w$  is within the area of  $0.3 \leq w < 1.0$ ) in the three component diagram defined by the apexes corresponding to each component.

In the dielectric ceramic composition described above, it is preferable that the second side component, when its composition is represented by  $x\text{SiO}_2 - y\text{TiO}_2 - z\text{XO}$  (wherein  $x$ ,  $y$  and  $z$  represent mol%), falls within or on the boundary lines of the area surrounded by straight lines connecting each point indicated by A ( $x = 85$ ,  $y = 1$ ,  $z = 14$ ), B ( $x = 35$ ,  $y = 51$ ,  $z = 14$ ), C ( $x = 30$ ,  $y = 20$ ,  $z = 50$ ) and D ( $x = 39$ ,  $y = 1$ ,  $z = 60$ ) in the three component diagram defined by the apexes corresponding to each component.

The second side component contains in total of 15 parts by weight of at least one of  $\text{Al}_2\text{O}_3$  and  $\text{ZrO}_2$  (the content of  $\text{ZrO}_2$  is 5 parts by weight or less) relative to 100 parts by weight of the oxide represented by  $\text{SiO}_2 - \text{TiO}_2 - \text{XO}$ .

The present invention according to a different aspect provides a monolithic ceramic capacitor provided with a plurality of dielectric ceramic layers, inner electrodes formed between the ceramic layers and outer electrodes being electrically connected to the inner electrodes, wherein the dielectric ceramic layers are constructed by the dielectric ceramic composition described above and the inner electrodes are composed of nickel or a nickel alloy.

The outer electrode may be provided with a sintered layer of an electroconductive metal powder or an electroconductive metal powder supplemented with glass frits.

#### BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a cross sectional view showing the monolithic ceramic capacitor according to one embodiment of the present invention.

FIG. 2 is a plane view showing the dielectric ceramic layer portion having inner electrodes of the monolithic ceramic capacitor shown in FIG. 1.

FIG. 3 is a disassembled perspective view showing the ceramic monolithic portion of the monolithic ceramic capacitor.

itor shown in FIG. 1.

FIG. 4 is a three component phase diagram of  $\text{Li}_2\text{O}-(\text{Si}_w, \text{Ti}_{1-w})\text{O}_2$ -MO oxides.

FIG. 5 is a three component phase diagram of  $\text{SiO}_2$ - $\text{TiO}_2$ -XO oxides.

## DESCRIPTION OF THE PREFERRED EMBODIMENT

The basic construction of the monolithic ceramic capacitor according to the first embodiment of the present invention will be described hereinafter referring to the drawings. FIG. 1 is a cross section showing one example of the monolithic ceramic capacitor, FIG. 2 is a plane view showing the dielectric ceramic portion having inner electrodes of the monolithic ceramic capacitor in FIG. 1, and FIG. 3 is a disassembled perspective view showing the dielectric ceramic portion having inner electrodes of the monolithic ceramic capacitor in FIG. 1.

As shown in FIG. 1, the monolithic ceramic capacitor 1 according to the present embodiment is provided with a rectangular shaped monolithic ceramic body 3 obtained by laminating a plurality of dielectric ceramic layers 2a and 2b via the inner electrodes 4. An outer electrode 5 is formed on the both side faces of the monolithic ceramic body 3 so that the outer electrodes are electrically connected to each of the specified inner electrodes 4, on which a first plating layer 6 comprising nickel or copper is plated, a second plating layer 7 comprising a solder or tin being further formed on the first plating layer.

The method for producing the monolithic ceramic capacitor 1 will be next described in the order of production steps.

At first, a raw material powder of barium titanate prepared by weighing and mixing in a given composition ratio is prepared as an essential component of the dielectric ceramic layers 2a and 2b.

Then, a slurry is prepared by adding an organic binder in this raw material powder and, after forming this slurry into a sheet, a green sheet for use in the dielectric ceramic layers 2a and 2b is obtained.

Next, an inner electrode 4 comprising nickel or a nickel alloy is formed on one principal face of the green sheet to serve as the dielectric ceramic layers 2b. Nickel or nickel alloys as base metals may be used for the material of the inner electrode 4 when the dielectric ceramic layers 2a and 2b are formed using the dielectric ceramic composition as described above. The inner electrode 4 may be formed by a screen printing method, a deposition method or a plating method.

After laminating a required number of the green sheets for use in the dielectric ceramic layers 2b having the inner electrodes 4, the green sheets are inserted between the green sheets for use in the dielectric ceramic layer 2a having no inner electrode, thus obtaining a raw monolithic body by press-adhering these green sheets.

Then, this raw monolithic body is fired at a given temperature to obtain a ceramic monolithic body 3.

The outer electrodes 5 are formed at the both side faces of the ceramic monolithic body 3 so as to be electrically connected to the inner electrodes 4. The same material as used in the inner electrodes 4 can be used for the outer electrodes 5. While silver, palladium, a silver-palladium alloy, copper and a copper alloy is available besides a composition prepared by adding a glass frit such as a  $\text{B}_2\text{O}_3$ - $\text{SiO}_2$ -BaO glass or  $\text{Li}_2\text{O}$ - $\text{SiO}_2$ -BaO glass into these metal powders, an appropriate material should be selected by taking the application object and application site of the monolithic capacitor into consideration. While the outer electrodes 5 is formed by coating the ceramic monolithic body 3 obtained by firing with a metal powder paste as a raw material followed by heat-adhering, it may be formed by heat-adhering the metal powder paste simultaneously with the ceramic monolithic body 3.

The first plating layer 6 is then formed by applying a plating with nickel or copper on the outer electrode 5. Finally, the second plating layer 7 comprising a solder or tin is formed on the first plating layer 6, thereby completing the monolithic capacitor 1. Such process for further forming a conductive layer on the outer electrode 5 may be omitted depending on the application field of the monolithic ceramic capacitor.

By using the dielectric ceramic composition as described previously for constructing the dielectric ceramic layers 2a and 2b, the characteristic of the dielectric ceramic layers is not deteriorated even when it is fired in a reducing atmosphere. In other words, such characteristics are obtained in which the product between the insulation resistance and the electrostatic capacitance (a product CR) is as high as  $4900$  to  $5000 \Omega \cdot \text{F}$  or more and  $200 \Omega \cdot \text{F}$  or more at room temperature and  $150^\circ\text{C}$ , respectively, when the capacitor is used under an electric field strength as high as about  $10 \text{ kV/mm}$ , along with having a small voltage dependence of the insulation resistance, the absolute value of the capacitance decreasing ratio at an impressed DC voltage of  $5 \text{ kV/mm}$  being as small as  $40\%$  to  $45\%$ , the insulation durability being as high as  $12 \text{ kV/mm}$  or more under an AC voltage and  $14 \text{ kV/mm}$  under a DC voltage, besides its temperature characteristics of the electrostatic capacitance satisfying the B-level characteristic standard stipulated in the JIS Standard in the temperature range of  $-25^\circ\text{C}$  to  $+85^\circ\text{C}$  and X7R-level characteristic standard stipulated in the EIA standard in the temperature range of  $-55^\circ\text{C}$  to  $+125^\circ\text{C}$  and being excellent in weather resistance performance shown by a high temperature load test at  $150^\circ\text{C}$  and at DC  $25 \text{ kV/mm}$  and high humidity load test.

It has been confirmed that, among alkali earth metal oxides such as SrO and CaO existing in barium titanate as impurities, alkali metal oxides such as  $\text{Na}_2\text{O}$  and  $\text{K}_2\text{O}$  and other oxides such as  $\text{Al}_2\text{O}_3$  and  $\text{SiO}_2$ , especially the content of the alkali metal oxides largely influences on the electric characteristics. While the specific dielectric constant is

decreased when the amounts of addition of rare earth element oxides such as  $\text{Eu}_2\text{O}_3$ ,  $\text{Gd}_2\text{O}_3$ ,  $\text{Tb}_2\text{O}_3$ ,  $\text{Dy}_2\text{O}_3$ ,  $\text{Ho}_2\text{O}_3$ ,  $\text{Er}_2\text{O}_3$ ,  $\text{Tm}_2\text{O}_3$  and  $\text{Yb}_2\text{O}_3$ , and  $\text{Sc}_2\text{O}_3$  and  $\text{Y}_2\text{O}_3$  are increased, the specific dielectric constant can be kept to a practically acceptable range of 900 to 1600 by keeping the content of the alkali metal oxides contained in barium titanate as impurities to 0.02% by weight or less.

5 Adding an oxide represented by  $\text{Li}_2\text{O}-(\text{Si}, \text{Ti})\text{O}_2-\text{MO}$  (wherein MO is at least one of  $\text{Al}_2\text{O}_3$  and  $\text{ZrO}_2$ ) in the dielectric ceramic composition allows the composition to be sintered at a relatively low temperature of 1300 °C or less, further improving the high temperature load characteristic.

10 Adding an oxide represented by  $\text{Si}_2\text{O}-\text{TiO}_2-\text{XO}$  (wherein XO is at least one kind of compound selected from  $\text{BaO}$ ,  $\text{CaO}$ ,  $\text{SrO}$ ,  $\text{MgO}$ ,  $\text{ZnO}$  and  $\text{MnO}$ ) in the dielectric ceramic composition allows the composition to be improved in sintering property as well as in high temperature load characteristic and humidity resistance load characteristic. A higher insulation resistance can be obtained by adding  $\text{Al}_2\text{O}_3$  and/or  $\text{ZrO}_2$  in the oxide represented by  $\text{Si}_2\text{O}-\text{TiO}_2-\text{XO}$ .

(Examples)

15 The present invention will now be described in more detail by way of examples. However, the embodiment within the scope of the present invention is not limited to these examples.

(Example 1)

20 After preparing and weighing  $\text{TiCl}_4$  and  $\text{Ba}(\text{NO}_3)_2$  having a variety of purity as starting materials, the compounds were precipitated as titanyl barium oxalate ( $\text{BaTiO}(\text{C}_2\text{O}_4) \cdot 4\text{H}_2\text{O}$ ) by adding oxalic acid. This precipitate was decomposed by heating at a temperature of 1000 °C or more to synthesize four kinds of barium titanate listed in TABLE 1.

Table 1

Kind of BaTiO <sub>3</sub>	Content of impurities (% by weight)					Mean particle size (μm)
	Alkali metal oxide	SrO	CaO	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	
A	0.003	0.012	0.001	0.01	0.005	0.6
B	0.02	0.01	0.003	0.019	0.008	0.56
C	0.012	0.179	0.018	0.155	0.071	0.72
D	0.062	0.014	0.001	0.019	0.004	0.58

Oxides, carbonates or hydroxides as each component of the first side component were weighed so as to be a composition ratio (mole ratio) of  $0.25\text{Li}_2\text{O}-0.65(0.30\text{TiO}_2+0.70\text{SiO}_2)-0.10\text{Al}_2\text{O}_3$  to obtain a powder by crushing and mixing. Likewise, oxides, carbonates or hydroxides as each component of the second side component were weighed so as

to be a composition ratio (mole ratio) of  $0.66\text{SiO}_2\text{-}0.17\text{TiO}_2\text{-}0.15\text{BaO}\text{-}0.02\text{MnO}$  to obtain a powder by crushing and mixing.

Oxide powders of the first and second side components were placed in separate platinum crucibles, respectively, and heated at  $1500^\circ\text{C}$ . After quenching and crushing the mixture, each oxide powder with a mean particle size of  $1\text{ }\mu\text{m}$  or less was obtained.

In the next step,  $\text{BaCO}_3$  for adjusting the mole ratio  $\text{Ba/Ti}$  ( $m$ ) in barium titanate,  $\text{Sc}_2\text{O}_3$ ,  $\text{Y}_2\text{O}_3$ ,  $\text{Eu}_2\text{O}_3$ ,  $\text{Gd}_2\text{O}_3$ ,  $\text{Tb}_2\text{O}_3$  and  $\text{Dy}_2\text{O}_3$ , and  $\text{BaZrO}_3$ ,  $\text{MgO}$  and  $\text{MnO}$ , each having a purity of 99% or more, were prepared. These raw material powders and the oxides described above to be either one of the side components were weighted so as to form compositions shown in TABLE 2 and TABLE 3. The amounts of addition of the first and second side components are indicated by parts by weight relative to 100 parts by weight of the essential component  $(\text{BaO})_m\text{TiO}_2 + \alpha\text{M}_2\text{O}_3 + \beta\text{R}_2\text{O}_3 + \gamma\text{BaZrO}_3 + \text{gMgO} + \text{hMnO}$ .

Table 2  
\* indicates "out of the scope of the present invention"

Sam- ple No.	(BaO) <sub>m</sub> · TiO <sub>2</sub> + αM <sub>2</sub> O <sub>3</sub> + β R <sub>2</sub> O <sub>3</sub> + γ BaZrO <sub>3</sub> + gMgO + hMnO	Amount of addition of the first side component											Amount of addition of the second side component		
		Kind of BaTiO <sub>3</sub>		Total of α					Total of β						
		α		β					γ	g	h	g + h		m	
		Sc <sub>2</sub> O <sub>3</sub>	Y <sub>2</sub> O <sub>3</sub>	Eu <sub>2</sub> O <sub>3</sub>	Gd <sub>2</sub> O <sub>3</sub>	Tb <sub>2</sub> O <sub>3</sub>	Dy <sub>2</sub> O <sub>3</sub>	α + β							
*1	A	0	0.0008	0	0.05	0	0	0.0508	0.02	0.05	0.07	0.12	1.005	1	0
*2	A	0.03	0.03	0	0	0.001	0	0.061	0.03	0.04	0.08	0.12	1.005	1	0
*3	A	0.01	0.01	0	0	0.0008	0	0.0208	0.03	0.02	0.03	0.05	1.005	1	0
*4	A	0	0.001	0	0.03	0.02	0.02	0.071	0.03	0.03	0.1	0.13	1.01	1	0
*5	A	0.01	0.02	0.02	0	0	0.02	0.07	0.03	0.12	0.01	0.13	1.01	1	0
*6	A	0.01	0.01	0	0.02	0.02	0	0.06	0	0.07	0.06	0.13	1.01	1.5	0
*7	A	0	0.01	0	0	0	0	0.02	0.03	0.03	0.04	0.07	1.01	1.5	0
*8	A	0.01	0.02	0	0	0.01	0.01	0.05	0.02	0.001	0.069	0.07	1.01	1	0
*9	A	0.01	0	0.01	0	0	0	0.02	0.02	0.125	0.005	0.13	1.01	1	0
*10	A	0.01	0.01	0.02	0.01	0	0	0.04	0.02	0.079	0.001	0.08	1.01	1	0
*11	A	0.01	0	0.01	0	0.01	0	0.03	0.02	0.005	0.13	0.13	1.01	1	0
*12	A	0.01	0.02	0.03	0.02	0	0	0.07	0.03	0.05	0.08	0.14	1.01	1	0
*13	A	0.005	0.005	0.01	0	0.01	0	0.02	0.03	0.03	0.025	0.055	0.99	1	0
*14	A	0.01	0	0.01	0.01	0.005	0.01	0.03	0.04	0.04	0.03	0.07	1.00	1	0
*15	A	0.005	0.005	0.01	0	0	0	0.01	0.04	0.02	0.03	0.05	1.038	1	0
*16	A	0.01	0	0.01	0	0	0.01	0.02	0.03	0.02	0.03	0.05	1.05	0	1
*17	A	0	0.01	0.01	0	0	0.01	0.02	0.03	0.03	0.04	0.07	1.01	0	0
*18	A	0	0.01	0.01	0	0.01	0.01	0.03	0.04	0.02	0.03	0.07	1.01	5	0
*19	A	0	0.01	0.01	0	0.01	0	0.02	0.03	0.02	0.04	0.06	1.01	0	0
*20	A	0.01	0.01	0.02	0	0	0.01	0.03	0.02	0.03	0.02	0.05	1.01	0	4
*21	D	0	0.01	0.01	0	0.01	0.02	0	0.03	0.04	0.05	0.09	1.01	2	0
22	A	0	0.001	0.001	0.02	0	0.009	0.03	0.02	0.04	0.01	0.05	1.015	1	0
23	B	0.01	0.01	0.02	0	0.01	0	0.01	0.03	0.03	0.02	0.05	1.02	1	0



Table 3

Sample No.	(BaO) <sub>m</sub> · TiO <sub>2</sub> + αMn <sub>2</sub> O <sub>3</sub> + βR <sub>2</sub> O <sub>3</sub> + γBaZrO <sub>3</sub> + gMgO + hMnO	Amount of addition of the first side component										Amount of addition of the second side component					
		Total of α				Total of β	α + β	γ	g	h	g + h		m				
		β															
		Sc <sub>2</sub> O <sub>3</sub>	Y <sub>2</sub> O <sub>3</sub>	Eu <sub>2</sub> O <sub>3</sub>	Gd <sub>2</sub> O <sub>3</sub>									Tb <sub>2</sub> O <sub>3</sub>	Dy <sub>2</sub> O <sub>3</sub>		
24	C	0.01	0.02	0.03	0	0	0	0.01	0.01	0.04	0.03	0.12	0.002	0.122	1.03	1	0
25	A	0.01	0.03	0.04	0	0	0.01	0	0.01	0.05	0.03	0.07	0.06	0.13	1.02	1	0
26	A	0.01	0.04	0.05	0	0.01	0	0	0.01	0.06	0.03	0.002	0.12	0.122	1.01	0	1
27	A	0.005	0.005	0.01	0	0	0	0.001	0.001	0.011	0.02	0.01	0.02	0.03	1.01	1	0
28	A	0	0.01	0.01	0.01	0.01	0	0	0.02	0.03	0.02	0.03	0.02	0.05	1.01	1	0
29	A	0	0.01	0.01	0.02	0	0.01	0	0.03	0.04	0.02	0.05	0.03	0.08	1.015	0	1
30	A	0	0.01	0.01	0	0.04	0	0	0.04	0.05	0.02	0.06	0.03	0.09	1.01	0	1
31	A	0	0.01	0.01	0	0.03	0	0.02	0.05	0.06	0.02	0.05	0.06	0.11	1.01	1	0
32	A	0.01	0.02	0.03	0	0	0.02	0	0.02	0.05	0.02	0.05	0.04	0.09	1.01	1	0
33	A	0.01	0.02	0.03	0	0	0	0.03	0.03	0.06	0.03	0.06	0.04	0.1	1.01	1	0
34	A	0	0.01	0.01	0	0.02	0	0	0.02	0.03	0.01	0.03	0.02	0.05	1.01	1	0
35	A	0.01	0.01	0.02	0.02	0	0	0	0.02	0.04	0.04	0.05	0.03	0.08	1.01	1	0
36	A	0.01	0.01	0.02	0	0.01	0.01	0	0.02	0.04	0.06	0.05	0.02	0.07	1.01	1	0
37	A	0.01	0.01	0.02	0	0	0.01	0.01	0.02	0.04	0.03	0.04	0.03	0.07	1.01	2	0
38	A	0.01	0.01	0.02	0.01	0	0	0	0.01	0.03	0.03	0.04	0.015	0.055	1.01	2	0
39	A	0.01	0.01	0.02	0	0.01	0	0	0.01	0.03	0.02	0.03	0.04	0.07	1.01	2	0
40	A	0	0.02	0.02	0	0	0.01	0	0.01	0.03	0.02	0.03	0.02	0.05	1.01	2	0
41	A	0	0.02	0.02	0	0	0	0.01	0.01	0.03	0.02	0.03	0.03	0.06	1.001	2	0
42	A	0.02	0	0.02	0.01	0.01	0	0	0.02	0.04	0.03	0.04	0.03	0.07	1.01	0	2
43	A	0.01	0.01	0.02	0	0	0.01	0.01	0.02	0.04	0.03	0.03	0.05	0.08	1.035	2	0
44	A	0.01	0.01	0.02	0	0.02	0	0	0.02	0.04	0.03	0.04	0.03	0.07	1.015	0.2	0
45	A	0.01	0.01	0.02	0	0	0	0.01	0.01	0.03	0.03	0.03	0.02	0.05	1.01	3	0
46	A	0	0.02	0.02	0	0	0	0.01	0.01	0.03	0.03	0.02	0.04	0.06	1.01	0	0.2
47	A	0	0.02	0.02	0.01	0	0.01	0	0.02	0.04	0.03	0.05	0.02	0.07	1.01	0	3

Organic solvents such as polyvinyl butyral binder and ethanol were added to the weighed compounds and the mixture was mixed in a ball mill in a wet state to prepare a ceramic slurry. This ceramic slurry was formed into a sheet by a doctor blade method to obtain a rectangular shaped green sheet with a thickness of 35 μm, followed by printing an

electroconductive paste mainly composed of Ni on the ceramic green sheet to form an electroconductive paste layer for forming inner electrodes.

Then, a plurality of the ceramic green sheets on which the electroconductive layer is formed were laminated so that the sides where the electroconductive paste is projected out are alternately placed with each other, thus obtaining a monolithic body. This monolithic body was heated at 350 °C in a N<sub>2</sub> atmosphere and, after allowing the binder to decompose, the monolithic body was fired at the temperatures shown in TABLE 4 and TABLE 5 in a reducing atmosphere comprising H<sub>2</sub>-N<sub>2</sub>-H<sub>2</sub>O gases under an oxygen partial pressure of 10<sup>-9</sup> to 10<sup>-12</sup> MPa, thereby obtaining a ceramic sintered body.

The both side faces of the ceramic sintered body were coated with a silver paste containing B<sub>2</sub>O<sub>3</sub>-Li<sub>2</sub>O-SiO<sub>2</sub>-BaO glass frits and fired at a temperature of 600 °C in a N<sub>2</sub> atmosphere, thereby obtaining outer electrodes electrically connected to the inner electrodes.

The overall dimensions of the monolithic ceramic capacitor thus obtained were 5.0 mm in width, 5.7 mm in length and 2.4 mm in thickness while the thickness of the dielectric ceramic layer was 30 μm. Total number of the effective dielectric ceramic layers were 57, the area of the confronting electrode per one layer being  $8.2 \times 10^{-6} \text{ m}^2$ .

Electric characteristics of these monolithic ceramic capacitors were measured. The electrostatic capacitance (C) and dielectric loss (tan δ) were measured using an automatic bridge type measuring instrument at 1 kHz, 1 Vrms and 25 °C and the dielectric constant (ε) was calculated from the electrostatic capacitance. Next, the insulation resistance was measured using an insulation resistance tester at 25 °C and 150 °C by impressing direct current voltages of 315 V (or 10 kV/mm) and 945 V (or 30 kV/mm) for 2 minutes, obtaining a product of the electrostatic capacitance and insulation resistance, or a product CR.

The rate of change of the electrostatic capacitance against temperature changes was also measured. The rate of change at -25 °C and 85 °C by taking the electrostatic capacitance at 20 °C as a standard ( $\Delta C/C_{20}$ ), the rate of change at -55 °C and 125 °C by taking the electrostatic capacitance at 25 °C as a standard ( $\Delta C/C_{25}$ ) and the maximum value of the rate of change ( $|\Delta C| \text{ max}$ ) as an absolute value were measured as the electrostatic capacitances against temperature changes.

The DC bias characteristic was also evaluated. First, the electrostatic capacitance when an AC voltage of 1 kHz and 1 Vrms was impressed was measured. Then, the electrostatic capacitance when a DC voltage of 150 V and an AC voltage of 1 kHz and 1 Vrms were simultaneously impressed was measured, thereby the rate of reduction of the electrostatic capacitance ( $\Delta C/C$ ) due to loading the DC voltage was calculated.

In the high temperature load test, a direct current voltage of 750 V (or 25 kV/mm) was impressed at 150 °C on 36 pieces of each sample to measure the time dependent changes of the insulation resistance. The time when the insulation resistance of each sample was reduced below 10<sup>6</sup> Ω was defined to be a life span time and mean life span time was evaluated.

In the humidity resistance test, the number of the test pieces having an insulation resistance of 10<sup>6</sup> Ω or less among the 72 test pieces were counted after impressing a DC voltage of 315 V under an atmospheric pressure of 2 atm (relative humidity 100%) at 120 °C for 250 hours.

Insulation breakdown voltages under AC and DC voltages were measured by impressing AC and DC voltages at a voltage increase rate of 100 V/sec.

The results described above are listed in TABLE 4 and TABLE 5.

Table 4  
 \* indicates "out of the scope of the present invention"

Sam- ple No.	Baking tempera- ture (°C)	Dielec- tric con- stant	Dielec- tric loss tan δ (%)	Ratio of temperature dependent capacitor change (%)				DC bias charac- teristic (%) ΔC/C 5kV/mm	Product CR (Ω · F)				Insulation breakdown voltage (kV/mm)		Humidity resistance load test; Number of rejects	Mean life span (h)	
				ΔC/C <sub>20</sub>		ΔC/C <sub>25</sub>			Maxi- mum value	315V Im- pressed Voltage		315 V Im- pressed Voltage					
				-25°C	85°C	-55°C	125°C			25°C		150°C					
										5110	4860	220	210	12			14
				25°C	150°C	25°C	150°C		25°C	150°C	25°C	150°C	25°C	150°C			25°C
*1	1300	1210	0.7	5.6	-12	6.4	-17.5	21	-21	5110	4860	220	210	12	14	07/2	960
*2	1300	960	0.7	2.3	-7.8	4.7	-6.9	8.7	-16	8520	8090	200	190	12	14	07/2	910
*3	1300	1550	0.7	3	-7.9	5	-6.8	8.5	-42	3020	2870	120	110	13	14	07/2	930
*4	1300	920	0.7	6	-12.9	7.5	-19	25.3	-14	5060	4810	250	240	12	14	07/2	120
*5	1280	960	2	2.1	-8	4.2	-7.1	8.5	-14	5070	4820	260	250	12	14	10/72	180
*6	1280	1070	0.7	1.9	-8.2	3	-7.5	8.9	-16	3120	2180	140	100	12	14	07/2	870
*7	1300	1440	0.7	2.2	-14.3	4.5	-31.5	36.2	-36	5160	4900	240	230	12	14	07/2	160
*8	1280	1280	0.8	2.2	-12.5	4.6	-16.3	21.3	-26	3090	2940	130	120	13	14	07/2	950
*9	1360	1530	2.6	2.3	-7.7	5	-7.5	8.5	-43	5110	4860	230	220	12	14	53/72	120
*10	Unmeasurable due to semiconductor formation																
*11	1280	1460	0.7	3	-8.5	5.1	-17.9	23.6	-38	3150	2990	150	140	12	14	07/2	150
*12	1280	940	2.1	2.3	-8.2	4.5	-8.5	8.7	-14	5060	4800	240	230	12	14	9/72	100
*13	Unmeasurable due to semiconductor formation																
*14	1300	1360	0.7	3.4	-8.4	5.3	-8	9.3	-30	3200	3040	160	150	10	11	07/2	130
*15	Unmeasurable due to insufficient sintering																
*16	Unmeasurable due to insufficient sintering																
*17	Unmeasurable due to insufficient sintering																
*18	1300	1320	2.6	3.3	-8.3	5.1	-8.2	9.2	-25	3250	3090	170	160	11	12	07/2	150
*19	Unmeasurable due to insufficient sintering 11																
*20	1300	1470	2.6	1.9	-8.7	4	-8	9.3	-41	3300	3140	180	170	12	12	07/2	110
*21	1300	1140	0.6	2.2	-8.9	7	-8.3	9.5	-26	5180	4920	280	270	12	14	07/2	860
22	1280	1480	0.7	5.2	-7.2	6.5	-7	8.7	-39	5090	4840	270	260	12	15	07/2	920
23	1280	1460	0.7	1.6	-7.6	7	-7.2	8.8	-39	5020	4770	250	240	12	14	07/2	940

Table 5  
\* indicates "out of the scope of the present invention"

Sam- ple No.	Baking tempera- ture (°C)	Dielec- tric con- stant	Dielec- tric loss tan δ (%)	Ratio of temperature dependent capaci- tance change (%)						DC bias charac- teristic (%)		Product CR (Ω · F)				Insulation breakdown voltage (kV/mm)		Humidity resistance load test; Number of reject	Mean life span (h)		
				ΔC/C <sub>20</sub>		ΔC/C <sub>25</sub>		Maxi- mum value	ΔC/C 5kV/mm	315V Im- pressed Voltage	945V Im- pressed Voltage	315 V Im- pressed Voltage		AC	DC						
				-25°C	85°C	-55°C	125°C					25°C	5020			290	280				
																				-31	290
-17	220	210	15	0/72	950																
						-45	230	220	14	0/72	820										
-36	280	270	14	0/72	850																
						-33	290	280	13	14	0/72	900									
-22	250	240	12	14	0/72								870								
						-15	210	200	12	14	0/72	920									
-21	200	190	12	14	0/72								920								
						-16	260	250	12	14	0/72	870									
-36	280	270	12	14	0/72								850								
						-30	230	220	12	15	0/72	820									
-32	220	210	12	14	0/72								850								
						-32	250	240	12	14	0/72	890									
-39	230	220	13	14	0/72								900								
						-39	240	230	13	14	0/72	870									
-40	210	200	12	14	0/72								910								
						-40	260	250	12	14	0/72	890									
-30	230	220	12	14	0/72								920								
						-30	250	240	12	14	0/72	900									
-25	230	220	12	14	0/72								880								
						-38	290	280	12	14	0/72	850									
-38	270	260	12	14	0/72								860								
						-38	260	250	12	14	0/72	920									

It is evident from Table 1 to TABLE 5 that the monolithic ceramic capacitor according to the present invention has a capacitance decreasing ratio of as small as within -45% at an impressed voltage of 5 kV/mm and a dielectric loss of less than 1.0 %, wherein the rate of change of the electrostatic capacitance against temperature changes satisfies both

the B-level characteristic standard stipulated in the JIS Standard in the temperature range of  $-25^{\circ}\text{C}$  to  $+85^{\circ}\text{C}$  and X7R-level characteristic standard stipulated in the EIA standard in the temperature range of  $-55^{\circ}\text{C}$  to  $+125^{\circ}\text{C}$ .

Moreover, the insulation resistances at  $25^{\circ}\text{C}$  and  $150^{\circ}\text{C}$  as expressed by the product CR show as high values as  $5000\ \Omega \cdot \text{F}$  or more and  $200\ \Omega \cdot \text{F}$  or more, respectively, when the ceramic capacitor is used under a high electric field strength of  $10\ \text{kV/mm}$ . The insulation breakdown voltage also shows high values of  $12\ \text{kV/mm}$  or more under the AC voltage and  $14\ \text{kV/mm}$  or more under the DC voltage. In addition, an acceleration test at  $150^{\circ}\text{C}$  and DC  $25\ \text{kV/mm}$  gave a mean life span as long as 800 hours or more besides enabling a relatively low firing temperature of  $1300^{\circ}\text{C}$  or less.

The reason why the composition was limited in the present invention will be described hereinafter.

In the composition of  $(\text{BaO})_m\text{TiO}_2 + \alpha\text{M}_2\text{O}_3 + \beta\text{R}_2\text{O}_3 + \gamma\text{BaZrO}_3 + \text{gMgO} + \text{hMnO}$  (wherein  $\text{M}_2\text{O}_3$  represents at least one of either  $\text{Sc}_2\text{O}_3$  or  $\text{Y}_2\text{O}_3$  and  $\text{R}_2\text{O}_3$  represents at least one of the compound selected from  $\text{Eu}_2\text{O}_3$ ,  $\text{Gd}_2\text{O}_3$ ,  $\text{Tb}_2\text{O}_3$  and  $\text{Dy}_2\text{O}_3$ ,  $\alpha$ ,  $\beta$ ,  $\gamma$ , g and h representing mole ratio, respectively), the  $\text{M}_2\text{O}_3$  content  $\alpha$  of less than 0.001 as shown in the sample No. 1 is not preferable because the temperature characteristic does not satisfy the B-level characteristic / X7R characteristic. On the other hand, the  $\text{M}_2\text{O}_3$  content  $\alpha$  of more than 0.05 as shown in the sample No. 2 is also not preferable because the specific dielectric constant is reduced to less than 1000. Accordingly, the preferable range of the  $\text{Mn}_2\text{O}_3$  content  $\alpha$  is  $0.001 \leq \alpha \leq 0.05$ .

It is not preferable that the  $\text{R}_2\text{O}_3$  content  $\beta$  is less than 0.001 as in the sample No. 3 since the insulation resistance is so low that the product CR becomes small. It is also not preferable that the  $\text{R}_2\text{O}_3$  content  $\beta$  is more than 0.05 as in the sample No. 4 because the temperature characteristic does not satisfy the B-level characteristic / X7R characteristic, reducing the reliability. Accordingly, the preferable range of the  $\text{R}_2\text{O}_3$  content  $\beta$  is  $0.001 \leq \beta \leq 0.05$ .

When the combined amount of  $\text{M}_2\text{O}_3$  and  $\text{R}_2\text{O}_3$  ( $\alpha + \beta$ ) is more than 0.06, the dielectric loss is increased up to 2.0% while the mean life span is shortened, being not preferable since the number of rejects in the humidity resistance load test is increased. Accordingly, the combined amount of  $\text{M}_2\text{O}_3$  and  $\text{R}_2\text{O}_3$  ( $\alpha + \beta$ ) is preferably in the range of  $\alpha + \beta \leq 0.06$ .

It is not preferable that, as seen in the sample No. 6, the  $\text{BaZrO}_3$  content  $\gamma$  is zero since the insulation resistance becomes low while having a larger voltage dependency of the insulation resistance than in the system containing  $\text{BaZrO}_3$ . On the other hand, when the  $\text{BaZrO}_3$  content  $\gamma$  exceeds 0.06 as in the sample No. 7, the temperature characteristic does not satisfy the B-level characteristic / X7R characteristic, being not preferable since the mean life span is shortened. Accordingly, the preferable range of the  $\text{BaZrO}_3$  content  $\gamma$  is  $0.005 \leq \gamma \leq 0.06$ .

It is not preferable that, as seen in the sample No. 8, the  $\text{MgO}$  content g is 0.001 since the insulation resistance becomes low besides the temperature characteristics does not satisfy the B-level characteristic / X7R characteristic. On the other hand, when the  $\text{MgO}$  content g exceeds 0.12 as seen in the sample No. 9, the sintering temperature becomes high and the dielectric loss exceeds 2.0%, which is not preferable because rejections in the humidity resistance test are extremely increased while shortening the mean life span. Accordingly, the preferable range of the  $\text{MgO}$  content g is  $0.001 \leq g \leq 0.12$ .

It is not preferable that the  $\text{MnO}$  content h is 0.001 as seen in the sample No. 10 since the sample becomes not measurable due to semiconductor formation. It is not preferable that the  $\text{MnO}$  content h exceeds 0.12, on the other hand, because the temperature characteristic X7R is not satisfied along with the insulation resistance becomes low and the mean life span becomes short. Accordingly, the preferable  $\text{MnO}$  content h is in the range of  $0.001 < h \leq 0.12$ .

It is not preferable that, as seen in the sample No. 12, the combined content of  $\text{MgO}$  and  $\text{MnO}$  ( $g + h$ ) exceeds 0.13 because the dielectric loss is increased to 2.0%, the mean life span is shortened and the number of rejects in the humidity resistance load test is increased. Accordingly, the combined content of  $\text{MgO}$  and  $\text{MnO}$  ( $g + h$ ) is preferably in the range of  $g + h \leq 0.13$ .

It is not preferable that the  $\text{BaO}/\text{TiO}_2$  ratio m is less than 1.000 as in the sample No. 13 because measurements are impossible due to formation of semiconductors. It is also not preferable that, as seen in the sample No. 14, that the  $\text{BaO}/\text{TiO}_2$  ratio m is 1.000 since the insulation resistance as well as the AC and DC breakdown voltage becomes low along with shortening the mean life span. It is not preferable, on the other hand, that the  $\text{BaO}/\text{TiO}_2$  ratio m is over 1.035 since measurements becomes impossible due to insufficient sintering. Accordingly, the  $\text{BaO}/\text{TiO}_2$  ratio m in the range of  $1.000 < m \leq 1.035$  is preferable.

It is not preferable that the amount of addition of the first or second side component is zero as in the samples No. 17 and 19 because measurements are impossible due to insufficient sintering. When the amount of addition of the first or second side component exceeds 3.0 parts by weight as seen in the samples No. 18 and 20, the dielectric loss exceeds 1.0% and the insulation resistance and insulation breakdown voltage are lowered along with shortening the mean life span, which are not preferable. Accordingly, the preferable content of either the first or the second components is 0.2 to 3.0 parts by weight.

The contents of the alkali metal oxides contained in barium titanate as impurities are suppressed below 0.02% by weight because, when the contents of the alkali earth metal oxides exceeds 0.02% by weight, the dielectric constant is decreased.

(Example 2)

A starting material  $\text{BaO}_{1.010} \cdot \text{TiO}_2 + 0.010\text{Y}_2\text{O}_3 + 0.02\text{Gd}_2\text{O}_3 + 0.01\text{BaZrO}_3 + 0.05\text{MgO} + 0.01 \text{ MnO}$  (mole ratio) was prepared using barium titanate "A" in TABLE 1 as a dielectric powder, in which  $\text{Li}_2\text{O} \cdot (\text{Si}, \text{Ti})\text{O}_2 \cdot \text{MO}$  oxide with a mean particle size of  $1 \mu\text{m}$  or less shown in TABLE 6 prepared by heating at 1200 to 1500 °C was added as a first side component. A monolithic ceramic capacitor was prepared by the same method as in Example 1, except that the starting material as described above was used. The overall dimensions of the monolithic ceramic capacitor produced are the same as in Example 1.

Table 6

Sample No.	Amount of addition (parts by weight)	The first side component				
		Composition (mol %, except w)				
		$\text{Li}_2\text{O}$	$(\text{Si}_w\text{Ti}_{1-w})\text{O}_2$	w	$\text{Al}_2\text{O}_3$	$\text{ZrO}_2$
101	1	20	80	0.3	0	0
102	1	10	80	0.6	5	5
103	0.8	10	70	0.5	20	0
104	0.8	35	45	1	10	10
105	1.5	45	45	0.5	10	0
106	1.5	45	55	0.3	0	0
107	1	20	70	0.6	5	5
108	1	20	70	0.4	10	0
109	1.2	30	60	0.7	5	5
110	1.2	30	60	0.8	10	0
111	2	40	50	0.6	5	5
112	2	40	50	0.9	0	10
113	1.5	10	85	0.4	5	0
114	2	5	75	0.6	10	10
115	1.2	20	55	0.5	25	0
116	1	45	40	0.8	0	15
117	0.8	50	45	0.7	5	0
118	1.2	25	75	0.9	0	0
119	1.5	25	75	1	0	0
120	1	35	65	0.9	0	0
121	1.5	35	65	1	0	0
122	1.2	20	70	0.2	0	10

Then, the electric characteristics were measured by the same method as described in example 1. The results are shown in TABLE 7.

Table 7

Sam- ple No.	Baking temp. (°C)	Dielec- tric con- stant	Dielec- tric loss tan δ (%)	Ratio of temperature dependent capaci- tance change (%)				DC bias charac- teristic (%) ΔC/C 5kV/mm	Product CR (Ω · F)				Insulation breakdown voltage (kV/mm)		Humidity resistance load test; Number of reject	Mean life span (h)	
				ΔC/C <sub>20</sub>		Maxi- mum value	315V Im- pressed Voltage		945V Im- pressed Voltage	315 V Im- pressed Voltage	945 V Im- pressed Voltage	AC	DC				
				ΔC/C <sub>25</sub>													
				-25°C	85°C									-55°C			125°C
101	1280	1480	0.6	2.1	-8.5	5.2	-8	8.3	-39	5090	4840	230	220	12	15	0/72	880
102	1280	1490	0.6	2.3	-8	5.6	-8.5	8.9	-40	5080	4830	240	230	12	14	0/72	850
103	1280	1420	0.6	3	-8.1	4.9	-8.9	9.5	-38	5070	4820	250	240	12	14	0/72	900
104	1300	1400	0.6	2.4	-8.3	5	-9.2	9.5	-36	5100	4850	230	220	12	15	0/72	910
105	1300	1460	0.6	2.6	-8	5.2	-9.5	9.7	-37	5120	4860	220	210	13	14	0/72	820
106	1280	1440	0.7	2.1	-8.9	4.8	-8.2	8.8	-37	5100	4850	230	220	12	14	0/72	820
107	1280	1500	0.6	2	-7.9	4.9	-9.1	9.5	-40	5230	4970	250	240	13	14	0/72	910
108	1280	1480	0.6	3.1	-7.8	5.2	-9.4	9.6	-40	5130	4870	240	230	12	14	0/72	930
109	1280	1480	0.6	2.8	-8.2	5.4	-9	9.2	-40	5090	4840	230	220	12	14	0/72	880
110	1300	1490	0.6	2.5	-8.2	5.5	-9.5	9.8	-40	5080	4830	220	220	12	14	0/72	860
111	1300	1460	0.6	2	-8.5	5.7	-9.1	9.5	-39	5070	4820	220	210	12	14	0/72	880
112	1280	1470	0.6	2.8	-8	5	-9	9.2	-39	5130	4870	220	210	12	14	0/72	870
113	1350	Unmeasurable due to insufficient sintering															
114	1350	Unmeasurable due to insufficient sintering															
115	1350	1450	1.4	2.2	-8.9	4.8	-8.7	9	-39	5160	4900	250	240	11	13	20/72	120
116	1350	Unmeasurable due to insufficient sintering															
117	1350	Unmeasurable due to insufficient sintering															
118	1300	1450	0.6	2.3	-8.8	5.3	-8.9	9.2	-39	5200	4940	240	230	12	14	0/72	820
119	1350	1490	1.3	1.9	-8.5	4.5	-8.6	8.9	-40	5190	4930	260	250	11	13	11/72	190
120	1300	1440	0.6	2.4	-9.2	5	-9.2	9.3	-37	5180	4920	250	240	12	14	0/72	860
121	1350	1460	1.3	2.1	-8.8	4.2	-8.7	9	-37	5170	4910	240	230	11	13	22/72	120
122	1350	1450	1.2	2.2	-8.7	4.5	-8.8	9.2	-37	5200	4940	230	220	11	13	19/72	170

As is evident from TABLE 6 and TABLE 7, preferable results are obtained in the samples No. 101 to 112, 118 and 120, in which oxides with compositions within or on the boundary lines of the area surrounded by the straight lines con-

necting each spot indicated by A ( $X = 20, y = 80, z = 0$ ), B ( $X = 10, y = 80, z = 10$ ), C ( $X = 10, y = 70, z = 20$ ), D ( $X = 35, y = 45, z = 20$ ), E ( $X = 45, y = 45, z = 10$ ) and F ( $X = 45, y = 55, z = 0$ ) in the three component phase diagram of the oxides represented by  $\text{Li}_2\text{O}-(\text{Si}_w, \text{Ti}_{1-w})\text{O}_2\text{-MO}$  shown in FIG. 4 are added, wherein the samples have a capacitance decreasing ratio of as small as within -45% at an impressed voltage of 5 kV/mm and a dielectric loss of 1.0% or less, along with the rate of change of the electrostatic capacitance against temperature changes satisfying the B-level characteristic standard stipulated in the JIS Standard in the temperature range of  $-25^\circ\text{C}$  to  $+85^\circ\text{C}$  and X7R-level characteristic standard stipulated in the EIA standard in the temperature range of  $-55^\circ\text{C}$  to  $+125^\circ\text{C}$ .

The insulation resistance represented by the product CR at  $25^\circ\text{C}$  and  $150^\circ\text{C}$  shows as high values as  $5000\ \Omega \cdot \text{F}$  or more and  $200\ \Omega \cdot \text{F}$  or more, respectively, when the capacitor used under a electric field strength of 10 kV/mm. The insulation breakdown voltages are as high as 12 kV/mm or more under AC voltage and 14 kV/mm or more under a DC voltage. The mean life span is as long as 800 hours or more in the acceleration test at  $150^\circ\text{C}$  and DC 25 kV/mm while enabling a relatively low firing temperature of  $1300^\circ\text{C}$  or less.

On the contrary, when the  $\text{Li}_2\text{O}-(\text{Si}_w, \text{Ti}_{1-w})\text{O}_2\text{-MO}$  oxides is outside of the composition range described above, the sintering becomes insufficient or many samples are rejected in the humidity resistance load test even after sintering in the samples No. 113 to 117 and 119. The samples with the composition falling on the line A-F and  $w = 1.0$  as in the samples No. 119 and 121, the sintering temperature becomes high along with causing many rejects in the humidity resistance load test. When the value of  $w$  is less than 0.30 as shown in the sample No. 122, the sintering temperature becomes high along with causing many rejects in the humidity resistance load test.

### (Example 3)

A starting material  $\text{BaO}_{1.010} \cdot \text{TiO}_2 + 0.010\text{Y}_2\text{O}_3 + 0.01\text{Eu}_2\text{O}_3 + 0.01\text{Gd}_2\text{O}_3 + 0.01\text{BaZrO}_3 + 0.05\text{MgO} + 0.01\text{MnO}$  (mole ratio) was prepared using barium titanate in TABLE 1A as a dielectric powder, in which oxides represented by  $\text{SiO}_2\text{-TiO}_2\text{-XO}$  with a mean particle size of  $1\ \mu\text{m}$  or less shown in TABLE 8 prepared by heating at  $1200$  to  $1500^\circ\text{C}$  was added as a second side component. A monolithic ceramic capacitor was produced by the same method as in Example 1, except that the starting material as described above was used. The amounts of addition of  $\text{Al}_2\text{O}_3$  and  $\text{ZrO}_2$  correspond to the amounts of addition relative to 100 parts by weight of the second side component ( $x\text{SiO}_2\text{-yTiO}_2\text{-zXO}$ ). The overall dimensions of the monolithic capacitor produced is the same as in Example 1.



Table 8

Sample No.	The second side component													
	Amount of addition (parts by weight)	Essential component (mole %)										Added component (parts by weight)		
		SiO <sub>2</sub>	TiO <sub>2</sub>	XO						Total				
				BaO	CaO	SrO	MgO	ZnO	MnO					
201	1	85	1	1	0	0	0	4	9	14	0	0		
202	1	35	51	0	10	0	0	0	4	14	0	0		
203	1	30	20	0	30	0	15	4	1	50	0	0		
204	1	39	1	20	20	2	0	13	5	60	0	0		
205	1	70	10	5	5	0	0	10	0	20	0	0		
206	1	45	10	0	0	0	0	15	30	45	0	0		
207	1	50	20	10	10	3	7	0	0	30	0	0		
208	1	50	30	0	16	0	0	0	4	20	0	0		
209	1	35	30	25	10	0	0	0	0	35	0	0		
210	1	40	40	10	0	0	0	5	5	20	0	0		
211	1	45	22	3	30	0	0	0	0	33	15	0		
212	1	45	22	3	30	0	0	0	0	33	10	5		
213	1	65	25	5	5	0	0	0	0	10	0	0		
214	1	25	40	15	0	10	0	5	5	35	0	0		
215	1	30	10	30	25	0	0	5	0	60	0	0		
216	1	50	0	35	15	0	0	0	0	50	0	0		
217	1	45	22	30	0	0	3	0	0	33	25	0		
218	1	45	22	30	0	3	0	0	0	33	0	15		
219	1	30	60	10	0	0	0	0	0	10	0	0		

Then, the electric characteristics were measured as in Example 1. The results are shown in TABLE 9.

Table 9

Sam- ple No.	Baking temp. (°C)	Dielec- tric con- stant	Dielec- tric loss tan δ (%)	Ratio of temperature dependent capaci- tance change (%)					DC vias charac- teristic (%) ΔC/C 5kV/mm	Product CR (Ω · F)				Insulation breakdown voltage (kV/mm)		Humidity resistance load test; Number of reject	Mean life span (h)	
				ΔC/C <sub>20</sub>			ΔC/C <sub>25</sub>			Maxi- mum value	315V Im- pressed Voltage	945V Im- pressed Voltage	315 V Im- pressed Voltage	945 V Im- pressed Voltage	AC			DC
				-25°C	85°C	-55°C	125°C											
201	1300	1460	0.6	3.2	-8.2	6.8	-7.9	8.5	-39	5080	4830	220	210	12	14	0/72	840	
202	1280	1490	0.6	3.5	-8.4	7	-8.1	8.6	-40	5080	4830	230	220	12	14	0/72	800	
203	1280	1470	0.6	4	-8.6	7.2	-8.3	8.7	-39	5120	4860	220	210	12	14	0/72	920	
204	1300	1450	0.6	3.8	-8.5	6.9	-8.2	8.6	-39	5150	4890	240	230	12	14	0/72	860	
205	1300	1460	0.6	3.9	-8.5	7.1	-8.2	8.8	-39	5070	4820	220	210	12	14	0/72	820	
206	1280	1430	0.6	3.7	-8.5	6.8	-8.3	8.7	-38	5080	4830	220	210	13	15	0/72	900	
207	1280	1450	0.6	3.5	-8.4	7	-8.1	8.7	-39	5030	4780	230	220	12	14	0/72	890	
208	1300	1470	0.6	3.1	-8	6.7	-7.9	8.3	-39	5040	4790	200	190	12	14	0/72	930	
209	1300	1460	0.6	3.5	-8.4	6.9	-8	8.7	-39	5080	4830	220	210	12	14	0/72	830	
210	1300	1450	0.6	3.9	-8.6	7.2	-8.2	8.8	-39	5100	4850	210	200	12	14	0/72	860	
211	1280	1430	0.6	4	-8.7	7.3	-8.5	8.9	-38	5410	5140	300	290	12	14	0/72	870	
212	1300	1440	0.6	3.8	-8.4	6.9	-8	8.7	-39	5420	5150	310	300	12	14	0/72	880	
213	1350	1420	1.2	3.1	-8.1	6.6	-7.6	8.3	-38	5120	4860	230	220	11	13	38/72	150	
214	1350									Unmeasurable due to insufficient sintering								
215	1350									Unmeasurable due to insufficient sintering								
216	1350	1420	1.3	3.3	-8.2	6.8	-7.8	8.5	-38	5030	4780	220	210	11	13	70/72	120	
217	1350									Unmeasurable due to insufficient sintering								
218	1350									Unmeasurable due to insufficient sintering								
219	1350									Unmeasurable due to insufficient sintering								

As is evident from TABLE 8 and TABLE 9, preferable results are obtained in the samples No. 201 to 212, in which oxides with compositions within or on the boundary lines of the area surrounded by the straight lines connecting each spot indicated by A ( $X = 85$ ,  $y = 1$ ,  $z = 14$ ), B ( $X = 35$ ,  $y = 51$ ,  $z = 14$ ), C ( $X = 30$ ,  $y = 20$ ,  $z = 50$ ) and D ( $X = 39$ ,  $y = 1$ ,  $z$

= 60), wherein x, y and z represent mole %, in the three component phase diagram of the  $\text{SiO}_2\text{-TiO}_2\text{-XO}$  oxides shown in FIG. 5 are added, wherein the samples have a capacitance decreasing ratio of as small as within -45% at an impressed voltage of 5 kV/mm and a dielectric loss of 1.0% or less, along with the rate of change of the electrostatic capacitance against temperature changes satisfying the B-level characteristic standard stipulated in the JIS Standard in the temperature range of -25 °C to +85 °C and X7R-level characteristic standard stipulated in the EIA standard in the temperature range of -55 °C to +125 °C.

When the capacitor is used under a high electric field strength of 10 kV/mm, the insulation resistance represented by the product CR at 25 °C and 150 °C show a high value of 5000  $\Omega \cdot \text{F}$  or more and 200  $\Omega \cdot \text{F}$  or more, respectively. The insulation breakdown voltage also shows high values of 12 kV/mm or more under the AC voltage and 14 kV/mm or more under the DC voltage. The mean life span in the acceleration test at 150 °C and DC 25 kV/mm is as long as 800 hours along with being free from rejects in the humidity resistance load test and being possible to sinter at a relatively low sintering temperature of 1300 °C.

In the  $\text{SiO}_2\text{-TiO}_2\text{-XO}$  oxide, on the other hand, sintering becomes insufficient as in samples No. 213 to 219 or rejects occur in the humidity resistance load test even after sintering.

Although a monolithic capacitor having 5400  $\Omega \cdot \text{F}$  or more and 300  $\Omega \cdot \text{F}$  or more of the insulation resistances at 25 °C and 150 °C, respectively, under the electric field strength of 10 kV/mm, the sintering property is extremely deteriorated when the amounts of addition of  $\text{Al}_2\text{O}_3$  and  $\text{ZrO}_2$  exceed 15 parts by weight and 5 parts by weight, respectively.

#### (Example 4)

Four kinds of barium titanate ( $\text{BaTiO}_3$ ) in TABLE 1, an oxide powder as a first side component and an oxide powder as a second side component were obtained by the same method as in Example 1.

Next,  $\text{BaCO}_3$  for adjusting the Ba/Ti mole ratio m of barium titanate, and  $\text{Sc}_2\text{O}_3$ ,  $\text{Y}_2\text{O}_3$ ,  $\text{Eu}_2\text{O}_3$ ,  $\text{Gd}_2\text{O}_3$ ,  $\text{Tb}_2\text{O}_3$  and  $\text{Dy}_2\text{O}_3$ , and  $\text{BaZrO}_3$  and  $\text{MnO}$ , each having a purity of 99% or more, were prepared. These raw material powders and the foregoing oxide powder as one of the side component of either the first or the second component were weighed to be the compositions shown in TABLE 10 and TABLE 11. The amounts of addition of the first and second side components correspond to the amount of addition relative to 100 parts by weight of the essential component  $(\text{BaO})_m\text{TiO}_2 + \alpha\text{M}_2\text{O}_3 + \beta\text{R}_2\text{O}_3 + \gamma\text{BaZrO}_3 + \text{gMnO}$ . A monolithic ceramic capacitor was produced using this weighed materials by the same method as in Example 1. Overall dimensions of the monolithic ceramic capacitor produced is the same as in Example 1.

Table 10

\* indicates "out of the scope of the present invention"

Sam- ple No.	(BaO) <sub>m</sub> · TiO <sub>2</sub> + α M <sub>2</sub> O <sub>3</sub> + β R <sub>2</sub> O <sub>3</sub> + γ BaZrO <sub>3</sub> + gMnO	indicates "out of the scope of the present invention"															The amount of addition of the first side com- ponent (parts by weight)	The amount of addition of the second side component (parts by weight)
		Kind of BaTiO <sub>3</sub>	α		Total of α	β				Total of β	α + β	γ	g	m				
			Sc <sub>2</sub> O <sub>3</sub>			Y <sub>2</sub> O <sub>3</sub>	Eu <sub>2</sub> O <sub>3</sub>	Gd <sub>2</sub> O <sub>3</sub>	Tb <sub>2</sub> O <sub>3</sub>						Dy <sub>2</sub> O <sub>3</sub>			
			Sc <sub>2</sub> O <sub>3</sub>	Y <sub>2</sub> O <sub>3</sub>														
*301	A	0	0.0007	0.0007	0.04	0	0	0	0.04	0.0407	0.03	0.09	0.01	1.5	0			
*302	A	0.05	0.02	0.07	0	0.001	0.001	0	0.002	0.072	0.02	0.14	1.005	1.2	0			
*303	A	0.02	0	0.02	0	0	0	0.0008	0.0008	0.0208	0.03	0.04	1.015	1	0			
*304	A	0	0.001	0.001	0.04	0	0.03	0	0.07	0.071	0.04	0.14	1.005	1	0			
*305	A	0	0.04	0.04	0	0.02	0.02	0	0.04	0.08	0.03	0.16	1.005	1	0			
*306	A	0.01	0	0.01	0.01	0	0	0.02	0.03	0.04	0	0.08	1.01	1.5	0			
*307	A	0	0.01	0.01	0	0	0.02	0	0.02	0.03	0.08	0.06	1.01	1.5	0			
*308	A	0.01	0.01	0.02	0	0.02	0	0	0.02	0.04	0.01	0.001	1.005	1	0			
*309	A	0	0.01	0.01	0.01	0	0	0	0.01	0.02	0.02	0.14	1.01	1	0			
*310	A	0.01	0	0.01	0	0	0	0.02	0.02	0.03	0.03	0.06	0.99	1.2	0			
*311	A	0	0.02	0.02	0	0.01	0.01	0.01	0.03	0.05	0.03	0.1	1.00	1	0			
*312	A	0.005	0.005	0.01	0	0.03	0	0	0.03	0.04	0.04	0.08	1.038	1	0			
*313	A	0	0.01	0.01	0	0	0	0.03	0.03	0.04	0.03	0.08	1.05	0	2			
*314	A	0.02	0	0.02	0.01	0.01	0	0	0.02	0.04	0.03	0.09	1.005	0	0			
*315	A	0	0.01	0.01	0.01	0	0	0.02	0.03	0.04	0.02	0.08	1.01	5	0			
*316	A	0.01	0	0.01	0	0	0.01	0.01	0.02	0.03	0.02	0.06	1.01	0	0			
*317	A	0.02	0	0.02	0.02	0	0	0	0.02	0.04	0.03	0.08	1.015	0	4			
*318	D	0.01	0	0.01	0	0	0	0.03	0.03	0.04	0.03	0.08	1.01	2.5	0			
319	A	0	0.001	0.001	0.01	0.01	0.01	0	0.03	0.031	0.02	0.06	1.015	1	0			
320	B	0	0.02	0.02	0	0	0	0.01	0.01	0.03	0.02	0.06	1.01	1	0			
321	C	0.01	0.01	0.02	0.01	0	0.02	0	0.03	0.05	0.03	0.11	1.01	1	0			
322	A	0.01	0.03	0.04	0	0	0	0.01	0.01	0.05	0.02	0.1	1.005	1	0			

Table 11

Sam- ple No.	(BaO) <sub>m</sub> · Kind of BaTiO <sub>3</sub>	(BaO) <sub>m</sub> · TiO <sub>2</sub> + α M <sub>2</sub> O <sub>3</sub> + β R <sub>2</sub> O <sub>3</sub> + γ BaZrO <sub>3</sub> + gMnO												The amount of addition of the second compo- nent (parts by weight)		
		α			Total of α	β				Total of β	α + β	γ	g		m	The amount of addition of the first component (parts by weight)
		Sc <sub>2</sub> O <sub>3</sub>		Y <sub>2</sub> O <sub>3</sub>		Eu <sub>2</sub> O <sub>3</sub>	Gd <sub>2</sub> O <sub>3</sub>	Tb <sub>2</sub> O <sub>3</sub>	Dy <sub>2</sub> O <sub>3</sub>							
		Sc <sub>2</sub> O <sub>3</sub>	Y <sub>2</sub> O <sub>3</sub>	Eu <sub>2</sub> O <sub>3</sub>		Gd <sub>2</sub> O <sub>3</sub>	Tb <sub>2</sub> O <sub>3</sub>	Dy <sub>2</sub> O <sub>3</sub>								
323	A	0	0.05	0.05	0.01	0	0	0	0.01	0.06	0.03	0.13	1.01	0	1	
324	A	0.02	0.01	0.03	0	0.001	0	0	0.001	0.031	0.02	0.06	1.005	1	0	
325	A	0.02	0	0.02	0	0	0	0.02	0.02	0.04	0.03	0.08	1.01	1.1	0	
326	A	0	0.01	0.01	0.01	0	0.02	0	0.03	0.04	0.02	0.08	1.01	0	1	
327	A	0.01	0	0.01	0	0	0.01	0.02	0.03	0.04	0.03	0.08	1.01	0	2	
328	A	0.01	0	0.01	0.03	0.02	0	0	0.05	0.06	0.02	0.12	1.01	1	0	
329	A	0.01	0.01	0.02	0.02	0	0	0	0.02	0.04	0.02	0.08	1.005	1	0	
330	A	0.01	0.02	0.03	0	0.01	0.01		0.02	0.05	0.02	0.1	1.01	1	0	
331	A	0.01	0	0.01	0	0.01	0	0.02	0.03	0.04	0.005	0.08	1.01	1	0	
332	A	0.01	0.01	0.02	0	0.02	0.01		0.03	0.05	0.03	0.1	1.01	1	0	
333	A	0.02	0	0.02	0.01	0	0	0.01	0.02	0.04	0.06	0.08	1.01	1	0	
334	A	0	0.02	0.02	0.02	0	0	0	0.02	0.04	0.03	0.08	1.005	2	0	
335	A	0.01	0.02	0.03		0	0.01	0	0.01	0.04	0.03	0.09	1.01	2	0	
336	A	0.01	0.01	0.02	0.01	0	0	0.01	0.02	0.04	0.03	0.08	1.01	2	0	
337	A	0.01	0.01	0.02	0.02	0.01	0	0	0.03	0.05	0.02	0.1	1.005	2	0	
338	A	0.005	0	0.005	0	0	0.005	0	0.005	0.01	0.02	0.02	1.001	2	0	
339	A	0.01	0.01	0.02	0	0	0.01	0.01	0.02	0.04	0.03	0.08	1.01	0	1.5	
340	A	0.03	0	0.03	0	0.02	0	0	0.02	0.05	0.02	0.1	1.035	2	0	
341	A	0	0.02	0.02	0	0	0.01	0.01	0.02	0.04	0.03	0.08	1.01	0.2	0	
342	A	0.01	0.01	0.02	0	0.01	0	0	0.01	0.03	0.03	0.06	1.01	3	0	
343	A	0.01	0.01	0.02	0.02	0	0.01	0	0.03	0.05	0.03	0.1	1.01	0	0.2	
344	A	0.02	0.01	0.03	0	0.03	0	0	0.03	0.06	0.02	0.13	1.005	0	3	

The electric characteristics were measured by the same method as in Example 1. The results are shown in TABLE 12 and TABLE 13.

Table 12

\* indicates "out of the scope of the present invention"

Sam- ple No.	Baking temp. (°C)	Dielec- tric con- stant	Dielec- tric loss tan δ (%)	Ratio of temperature dependent capacitance change (%)					DC bias charac- teristic (%) ΔC/C 5kV/mm	Product CR (Ω · F)				Insulation breakdown voltage (kV/mm)		Humidity resis- tance load test: Number of reject	Mean life span (h)
				ΔC/C <sub>20</sub>						315V Imt pressed Voltage	945V Imt pressed Voltage	315 V Imt pressed Voltage	945 V Imt pressed Voltage	AC	DC		
				ΔC/C <sub>25</sub>													
				ΔC/C <sub>25</sub>													
				-25°C	85°C	-55°C	125°C	Maxi- mum value									
*301	1300	1220	0.6	5.3	-12.5	6.5	-17.5	17.5	-18	5000	4750	210	200	13	14	0/72	980
*302	1300	820	0.7	2.5	-7.2	5	-7.5	7.6	-5	5110	4860	220	210	12	14	0/72	950
*303	1300	1450	0.6	3.2	-7.1	5.2	-7.6	8	-38	2900	2760	130	120	13	14	0/72	900
*304	1300	800	0.7	6.4	-13.5	7.6	-19.5	19.5	-7	4920	4670	230	220	12	14	0/72	100
*305	1280	710	2.2	3	-6.5	5	-7.3	7.5	-6	5030	4780	240	230	12	14	15/72	150
*306	1280	1220	0.6	2.2	-7.5	4	-8.4	8.6	-22	2850	2000	110	80	12	14	0/72	900
*307	1300	1340	0.7	2.5	-13.5	5	-31.5	31.5	-32	5110	4860	250	240	12	14	0/72	130
*308	Unmeasurable due to semiconductor formation																
*309	1280	1460	0.7	3.3	-7.5	5.5	-18.5	18.5	-37	2940	2790	160	150	12	14	0/72	110
*310	Unmeasurable due to semiconductor formation																
*311	1300	1160	0.7	4	-7.6	5.6	-8	8.3	-20	3080	2930	170	160	10	11	0/72	100
*312	Unmeasurable due to insufficient sintering																
*313	Unmeasurable due to insufficient sintering																
*314	Unmeasurable due to insufficient sintering																
*315	1300	1240	2.7	4	-7.4	5.8	-7.7	8.3	-22	3100	2950	160	150	10	12	0/72	130
*316	Unmeasurable due to insufficient sintering																
*317	1300	1260	2.8	2.3	-8.3	5	-8.5	8.8	-22	3110	2960	170	160	11	12	0/72	100
*318	1300	1250	0.7	2.5	-8.1	7.2	-8.2	8.8	-21	5050	4800	240	230	13	14	0/72	880
319	1280	1380	0.7	5.5	-6.6	7	-7.1	8.1	-35	5000	4750	240	230	12	15	0/72	960
320	1280	1360	0.6	1.7	-7.3	7.5	-8	8.2	-35	4990	4740	230	220	12	14	0/72	940
321	1280	1130	0.6	2	-8.1	4	-8.5	8.6	-18	5170	4910	250	240	12	15	0/72	970
322	1300	1130	0.6	2.2	-8.3	4.5	-8.5	8.7	-18	5090	4840	230	220	12	14	0/72	900

Table 13

Sam- ple No.	Baking temp. (°C)	Dielec- tric con- stant	Dielec- tric loss tan δ (%)	Ratio of temperature dependent capacitance change (%)				DC bias charac- teristic (%) ΔC/C 5kV/mm	Product CR (Ω · F)				Insulation breakdown voltage (kV/mm)		Humidity resis- tance load test: Number of reject	Mean life span (h)
				ΔC/C <sub>20</sub>		ΔC/C <sub>25</sub>			315V Im- pressed Voltage	945V Im- pressed Voltage	315 V Im- pressed Voltage	945 V Im- pressed Voltage	AC	DC		
				-25°C	85°C	-55°C	125°C									
323	1300	940	0.7	2.3	-8.2	5	-8.5	8.8	5000	4750	210	200	12	15	072	930
324	1300	1320	0.6	2.1	-7.5	5.5	-8	8.2	5120	4860	250	240	12	14	072	850
325	1300	1230	0.6	3.3	-7.5	6	-8.4	8.4	5170	4910	250	240	12	14	072	870
326	1280	1280	0.6	3	-7.8	5.5	-8.3	8.5	5080	4830	250	240	13	14	072	890
327	1280	1240	0.7	2.5	-8.1	5.1	-8.2	8.7	5120	4860	250	240	12	15	072	900
328	1300	930	0.6	2.5	-8.3	6	-8.5	8.8	5100	4850	250	240	13	14	072	960
329	1300	1250	0.6	2.3	-7.5	5.2	-7.9	8.5	5010	4760	220	210	12	14	072	990
330	1300	1110	0.7	3	-7.7	5	-8	7.9	5000	4750	220	210	12	14	072	900
331	1280	1260	0.6	2	-8	5.1	-8.2	8.1	4980	4730	220	210	12	15	072	880
332	1280	1160	0.6	2.4	-7.4	5	-8.1	8	5090	4840	250	240	12	14	072	820
333	1300	1220	0.7	3	-7.5	6.2	-8	8.1	4920	4670	220	210	12	14	072	870
334	1300	1240	0.7	2.1	-7.7	6	-8.5	8.1	5000	4750	200	190	12	14	072	900
335	1300	1250	0.6	1.9	-7.6	5.4	-8.3	8.5	5010	4760	200	190	13	14	072	920
336	1280	1260	0.7	2.6	-7.8	5.8	-7.9	8.2	5110	4860	250	240	13	14	072	880
337	1280	1150	0.6	2.3	-8.3	6.2	-8.5	8.8	5100	4850	250	240	12	14	072	960
338	1280	1520	0.6	2	-7.5	6.1	-8.1	8.2	5080	4830	250	240	12	14	072	900
339	1300	1260	0.6	2.4	-7.4	6.2	-7.5	8.3	4990	4740	200	190	12	14	072	950
340	1300	1110	0.7	2	-7.6	6	-7.9	8.1	5120	4860	250	240	12	14	072	940
341	1300	1270	0.6	2.6	-7.9	5.8	-8.5	8.5	5100	4850	250	240	12	14	072	900
342	1300	1350	0.7	2.4	-7.8	7	-8.4	8.5	5140	4880	250	240	12	14	072	920
343	1280	1130	0.7	2.2	-7.7	6.4	-8.3	8.5	5190	4930	250	240	12	14	072	900
344	1300	980	0.7	2	-8.1	6	-8.5	8.8	5100	4850	240	230	12	14	072	960

As is evident from TABLE 10 to TABLE 13, the monolithic capacitor according to the present invention has a capacitance decreasing ratio of as small as within -45% at an impressed voltage of 5 kV/mm and a dielectric loss of 1.0% or less, along with the rate of change of the electrostatic capacitance against temperature changes satisfying the B-level

characteristic standard stipulated in the JIS Standard in the temperature range of -25 °C to +85 °C and X7R-level characteristic standard stipulated in the EIA standard in the temperature range of - 55 °C to +125 °C.

Moreover, when the capacitor is used under a high electric field strength of 10 kV/mm, the insulation resistance represented by the product CR at 25°C and 150°C shows a high value of 4900 Ω · F or more and 200 Ω · F or more, respectively. The insulation breakdown voltage also shows high values of 12 kV/mm or more under the AC voltage and 14 kV/mm under the DC voltage. The mean life span in the acceleration test at 150°C and DC 25 kV/mm is as long as 800 hours along with being possible to sinter at a relatively low sintering temperature of 1300°C.

The reason why the composition was limited in the present invention will be described hereinafter.

In the composition of  $(\text{BaO})_m\text{TiO}_2 + \alpha\text{M}_2\text{O}_3 + \beta\text{R}_2\text{O}_3 + \gamma\text{BaZrO}_3 + \text{gMnO}$  (wherein  $\text{M}_2\text{O}_3$  represents at least one of either  $\text{Sc}_2\text{O}_3$  or  $\text{Y}_2\text{O}_3$  and  $\text{R}_2\text{O}_3$  represents at least one of the compound selected from  $\text{Eu}_2\text{O}_3$ ,  $\text{Gd}_2\text{O}_3$ ,  $\text{Tb}_2\text{O}_3$  and  $\text{Dy}_2\text{O}_3$ ,  $\alpha$ ,  $\beta$ ,  $\gamma$  and  $g$  representing mole ratio, respectively), the  $\text{M}_2\text{O}_3$  content  $\alpha$  of less than 0.001 as shown in the sample No. 301 is not preferable because the temperature characteristic does not satisfy the B-level characteristic / X7R characteristic. On the other hand, the  $\text{M}_2\text{O}_3$  content  $\alpha$  of more than 0.05 as shown in the sample No. 302 is also not preferable because the specific dielectric constant is reduced to less than 900. Accordingly, the preferable range of the  $\text{M}_2\text{O}_3$  content  $\alpha$  is  $0.001 \leq \alpha \leq 0.05$ .

It is not preferable that the  $\text{R}_2\text{O}_3$  content  $\beta$  of less than 0.001 as in the sample No. 303 since the insulation resistance is so low that the product CR becomes small. It is also not preferable that the  $\text{R}_2\text{O}_3$  content  $\beta$  is more than 0.05 as in the sample No. 304 because the temperature characteristic does not satisfy the B-level characteristic / X7R characteristic, reducing the reliability. Accordingly, the preferable range of the  $\text{R}_2\text{O}_3$  content  $\beta$  is  $0.001 \leq \beta \leq 0.05$ .

When the combined amount of  $\text{M}_2\text{O}_3$  and  $\text{R}_2\text{O}_3$  ( $\alpha + \beta$ ) is more than 0.06 as in the sample No. 305, the dielectric loss is increased up to 2.0% while the mean life span is shortened, being not preferable since the number of rejects in the humidity resistance load test is increased. Accordingly, the combined amount of  $\text{M}_2\text{O}_3$  and  $\text{R}_2\text{O}_3$  ( $\alpha + \beta$ ) is preferably in the range of  $\alpha + \beta \leq 0.06$ .

It is not preferable that, as seen in the sample No. 306, the  $\text{BaZrO}_3$  content  $\gamma$  is zero since the insulation resistance becomes low while having a larger voltage dependency of the insulation resistance than in the system containing  $\text{BaZrO}_3$ . On the other hand, when the  $\text{BaZrO}_3$  content  $\gamma$  exceeds 0.06 as in the sample No. 307, the temperature characteristic does not satisfy the B-level characteristic / X7R characteristic, being not preferable since the mean life span is shortened. Accordingly, the preferable range of the  $\text{BaZrO}_3$  content  $\gamma$  is  $0.005 \leq \gamma \leq 0.06$ .

It is not preferable that, as seen in the sample No. 308, the MnO content  $g$  is 0.001 since measurements are impossible due to semiconductor formation. On the other hand, it is not preferable that the MnO content  $g$  exceeds 0.13 as seen in the sample No. 309 because the temperature characteristic X7R is not satisfied besides the insulating resistance is low and the mean life span becomes short. Accordingly, the preferable range of the MnO content  $g$  is  $0.001 < g \leq 0.13$ .

It is not preferable that the  $\text{BaO}/\text{TiO}_2$  ratio  $m$  is less than 1.000 as in the sample No. 310 because measurements are impossible due to formation of semiconductors. It is also not preferable that, as seen in the sample No. 311, the  $\text{BaO}/\text{TiO}_2$  ratio  $m$  is 1.000 since the insulation resistance as well as the AC and DC breakdown voltage becomes low along with shortening the mean life span. It is not preferable, on the other hand, that the  $\text{BaO}/\text{TiO}_2$  ratio  $m$  is over 1.035 as in the samples No. 312 and 313 since measurements becomes impossible due to insufficient sintering. Accordingly, the  $\text{BaO}/\text{TiO}_2$  ratio  $m$  in the range of  $1.000 < m \leq 1.035$  is preferable.

It is not preferable that the amount of addition of the first or second side component is zero as in the samples No. 314 and 316 because measurements are impossible due to insufficient sintering. It is not preferable that the amount of addition of the first or second side component exceeds 3.0 parts by weight as seen in the samples No. 315 and 317, the dielectric loss exceeds 1.0% and the insulation resistance and insulation breakdown voltage are lowered along with shortening the mean life span. Accordingly, the preferable content of either the first or the second components is 0.2 to 3.0 parts by weight.

The contents of the alkali earth metal oxides contained in barium titanate as impurities are suppressed below 0.02% by weight because, when the contents of the alkali earth metal oxides exceeds 0.02% by weight as in the sample No. 318, the dielectric constant is decreased.

#### (Example 5)

A starting material of the composition of  $\text{BaO}_{1.010} \cdot \text{TiO}_2 + 0.01\text{Y}_2\text{O}_3 + 0.02\text{Dy}_2\text{O}_3 + 0.01\text{BaZrO}_3 + 0.06\text{MnO}$  (mole ration) was prepared using barium titanate in TABLE 1A as a dielectric powder, in which oxides represented by  $\text{Li}_2\text{O}$ -(Si, Ti) $\text{O}_2$ -MO with a mean particle size of 1 μm or less shown in TABLE 6 prepared by heating at 1200 to 1500 °C was added as a first side component. A monolithic ceramic capacitor was produced by the same method as in Example 1, except that the starting material as described above was used. The overall dimensions of the monolithic capacitor is the same as in Example 1. The electric characteristics were measured by the same method as in Example 1. The results are shown in TABLE 14. The samples No. 401 to 422 in TABLE 14 correspond to the samples No. 101 to 122 in TABLE



6, respectively, wherein, for example, the side component in the sample No. 101 in TABLE 6 is added to the sample No. 401 in TABLE 14.

Table 14

Sam- ple No.	Baking temp. (°C)	Dielec- tric con- stant	Dielec- tric loss tan δ (%)	Ratio of temperature dependent capacitance change (%)				DC bias charac- teristic (%) ΔC/C 5kV/mm	Product CR (Ω · F)				Insulation breakdown voltage (kV/mm)		Humidity resistance load test: Number of rejection.	Mean life span (h)	
				ΔC/C <sub>20</sub>					315V Im- pressed Voltage	945V Im- pressed Voltage	315 V Im- pressed Voltage	945 V Im- pressed Voltage	AC	DC			
				ΔC/C <sub>25</sub>													
				-25°C	85°C	-55°C	125°C										
401	1280	1350	0.7	2.2	-8.4	4.8	-8.2	8.4	-33	4950	4700	220	210	13	15	0/72	920
402	1280	1380	0.6	2.1	-8	4.9	-8.1	8.3	-36	4970	4720	240	230	12	14	0/72	940
403	1280	1360	0.6	2.7	-7.9	5	-7.8	8.1	-33	4960	4710	230	220	13	15	0/72	930
404	1300	1310	0.6	2.1	-7.7	5.1	-7.9	8.3	-31	4990	4740	250	240	12	14	0/72	900
405	1300	1380	0.6	2.3	-7.9	4.7	-8.4	8.6	-36	5040	4790	230	220	12	14	0/72	870
406	1280	1380	0.6	2	-7.7	4.7	-8.5	8.7	-36	5010	4760	240	230	12	14	0/72	950
407	1280	1370	0.6	2	-7.5	5	-8.5	8.8	-35	5140	4880	250	240	12	14	0/72	930
408	1280	1380	0.6	2.8	-8	5	-8.2	8.5	-36	5030	4780	250	240	12	14	0/72	940
409	1280	1380	0.6	3	-7.4	4.9	-8.4	8.3	-36	4980	4730	240	230	12	14	0/72	900
410	1300	1350	0.6	2.4	-7.6	5.1	-8.5	8.7	-33	5000	4750	240	230	12	14	0/72	970
411	1300	1320	0.7	2.3	-7.5	5.4	-8.3	8.5	-32	4990	4740	230	220	12	14	0/72	920
412	1280	1360	0.6	2.7	-7.5	5.1	-8.2	8.3	-33	5050	4800	210	200	12	14	0/72	900
413	1350								Unmeasurable due to insufficient sintering								
414	1350								Unmeasurable due to insufficient sintering								
415	1350	1350	1.4	2.4	-7.7	5	8.5	8.6	-33	5040	4790	240	230	11	13	32/72	110
416	1350								Unmeasurable due to insufficient sintering								
417	1350								Unmeasurable due to insufficient sintering								
418	1300	1350	0.6	2.5	-7.5	5.1	-8.3	8.5	-33	5080	4830	230	220	12	14	0/72	880
419	1350	1370	1.3	2	-7.3	4.9	-8.2	8.5	-35	5100	4850	250	240	11	13	25/72	140
420	1300	1350	0.7	2.4	-7.8	4.7	-8.5	8.7	-33	4960	4710	260	250	12	14	0/72	920
421	1350	1360	1.3	2.5	-8	4.3	-8.3	8.5	-33	5070	4820	250	240	11	13	33/72	110
422	1350	1330	1.2	2.3	-8.1	4.9	-8.2	8.5	-32	5070	4820	250	240	11	13	28/72	150

As is evident from TABLE 14, preferable results are obtained in the samples No. 401 to 412, 418 and 420, in which

oxides in the samples No. 101 to 112, 118 and 120 in TABLE 6 with compositions within or on the boundary line of the area surrounded by the straight lines connecting each spot indicated by A ( $X = 20, y = 80, z = 0$ ), B ( $X = 10, y = 80, z = 10$ ), C ( $X = 10, y = 70, z = 20$ ), D ( $X = 35, y = 45, z = 20$ ), E ( $X = 45, y = 45, z = 10$ ) and F ( $X = 45, y = 55, z = 0$ ), wherein x, y and z represent mole % and w is mole ratio, in the three component phase diagram of the  $\text{Li}_2\text{O}-(\text{Si}_w, \text{Ti}_{1-w})\text{O}_2$ -MO oxides shown in FIG. 4 are added, wherein the samples have a capacitance decreasing ratio of as small as within -40% at an impressed voltage of 5 kV/mm and a dielectric loss of 1.0% or less, along with the rate of change of the electrostatic capacitance against temperature changes satisfying the B-level characteristic standard stipulated in the JIS Standard in the temperature range of -25 °C to +85 °C and X7R-level characteristic standard stipulated in the EIA standard in the temperature range of -55 °C to +125 °C.

When the capacitor is used under a high electric field strength of 10 kV/mm, the insulation resistance represented by the product CR at 25 °C and 150 °C show a high value of 4900  $\Omega \cdot \text{F}$  or more and 200  $\Omega \cdot \text{F}$  or more, respectively. The insulation breakdown voltage also shows high values of 12 kV/mm or more under the AC voltage and 14 kV/mm under the DC voltage. The mean life span in the acceleration test at 150 °C and DC 25 kV/mm is as long as 800 hours along with being possible to sinter at a relatively low sintering temperature of 1300 °C.

On the contrary, when the oxides represented by  $\text{Li}_2\text{O}-(\text{Si}_w, \text{Ti}_{1-w})\text{O}_2$ -MO is outside of the composition range described above as in the samples No. 113 to 117 and 119 in TABLE 6, the sintering becomes insufficient or many samples are rejected in the humidity resistance load test even after sintering as seen in the samples No. 413 to 417 and 419. The samples with the composition falling on the line A-F and  $W = 1.0$  have high sintering temperature along with causing many rejects in the humidity resistance load test as seen in the samples No. 119 and 121 in TABLE 14. When the value of w is less than 0.30 as shown in the sample No. 122 in TABLE 6, the sintering temperature becomes high along with causing many rejects in the humidity resistance load test as seen in the sample NO. 433 in TABLE 14.

#### (Example 6)

A starting material with a composition of  $\text{BaO}_{1.010} \cdot \text{TiO}_2 + 0.01\text{Y}_2\text{O}_3 + 0.01\text{Eu}_2\text{O}_3 + 0.01\text{Tb}_2\text{O}_3 + 0.015\text{BaZrO}_3 + 0.06\text{MnO}$  (mole ration) was prepared using barium titanate in TABLE 1A as a dielectric powder, in which the oxide represented by  $\text{SiO}_2$ - $\text{TiO}_2$ -XO with a mean particle size of 1  $\mu\text{m}$  or less as shown in TABLE 8 prepared by heating at 1200 to 1500 °C was added as a second side component. A monolithic ceramic capacitor was produced by the same method as in Example 1, except that the starting material as described above was used. The overall dimensions of the monolithic capacitor produced is the same as in Example 1. The electric characteristics were measured by the same method as in Example 1. The results are shown in TABLE 15. The samples No. 501 to 519 in TABLE 15 correspond to the samples No. 201 to 219 in TABLE 8, respectively, wherein, for example, the sample No. 501 in TABLE 15 was obtained by adding the side component in the sample No. 201 in TABLE 8.

Table 15

Sam- ple No.	Baking temp. (°C)	Dielec- tric con- stant	Dielec- tric loss tan δ (%)	Ratio of temperature dependent capacitance change (%)				DC bias charac- teristic (%) 5kV/mm	Product CR (Ω · F)				Insulation breakdown voltage (kV/mm)		Humidity resis- tance load test: Number of reject	Mean life span (h)	
				ΔC/C <sub>20</sub>		ΔC/C <sub>25</sub>			Maxi- mum value	315V Im- pressed Voltage	945V Im- pressed Voltage	315V Im- pressed Voltage	945V Im- pressed Voltage	AC			DC
				-25°C	85°C	-55°C	125°C										
501	1300	1350	0.6	3.3	-8	6.2	-7.5	7.8	-34	4970	4720	210	200	12	14	0/72	900
502	1280	1370	0.6	3.2	-8.1	6.5	-7.7	7.9	-36	4980	4730	220	210	12	14	0/72	890
503	1280	1360	0.7	3.4	-8.2	6.7	-8	8.2	-34	5030	4780	220	210	12	14	0/72	930
504	1300	1330	0.6	4	-8	6.5	-8.1	8.2	-33	5020	4770	230	220	13	15	0/72	870
505	1300	1350	0.6	3.5	-8.1	6.8	-7.9	8.1	-34	4990	4740	230	220	12	14	0/72	850
506	1280	1320	0.6	3.4	-7.9	6.3	-7.7	8	-33	4980	4730	210	200	13	15	0/72	900
507	1280	1350	0.6	3.5	-8.1	6.4	-7.5	7.7	-34	4950	4700	230	220	12	14	0/72	880
508	1300	1360	0.6	3.3	-7.7	6.8	-7.8	8	-34	4950	4700	200	190	12	14	0/72	910
509	1300	1370	0.6	3.7	-8.1	6.5	-7.7	7.8	-36	4980	4730	230	220	12	14	0/72	880
510	1300	1360	0.6	3.5	-7.9	6.8	-7.5	7.7	-34	5000	4750	220	210	12	14	0/72	870
511	1280	1340	0.6	3.8	-8	7	-7.6	7.8	-33	5320	5050	310	300	12	14	0/72	900
512	1300	1330	0.6	3.4	-7.8	6.8	-7.8	8.2	-34	5330	5060	320	300	13	15	0/72	900
513	1350	1310	1.3	3.3	-7.8	6.8	-7.9	8.3	-33	5100	4850	220	210	11	13	42/72	120
514	1350								Unmeasurable due to insufficient sintering								
515	1350								Unmeasurable due to insufficient sintering								
516	1350	1300	1.2	3.5	-8	6.5	-7.5	7.7	-33	4950	4700	230	220	11	13	71/72	130
517	1350								Unmeasurable due to insufficient sintering								
518	1350								Unmeasurable due to insufficient sintering								
519	1350								Unmeasurable due to insufficient sintering								

As is evident from the samples No. 501 to 512 in TABLE 15, preferable results are obtained in the samples in which the oxides of the samples No. 201 to 212 in TABLE 8 with compositions within or on the boundary lines of the area sur-

rounded by the straight lines connecting each spot indicated by A ( $X = 85$ ,  $y = 1$ ,  $z = 14$ ), B ( $X = 35$ ,  $y = 51$ ,  $z = 14$ ), C ( $X = 30$ ,  $y = 20$ ,  $z = 50$ ) and D ( $X = 39$ ,  $y = 1$ ,  $z = 60$ ), wherein  $x$ ,  $y$  and  $z$  represent mole %, in the three component phase diagram of the oxides represented by  $\text{SiO}_2\text{-TiO}_2\text{-XO}$  shown in FIG. 5 are added, wherein the samples have a capacitance decreasing ratio of as small as within -40% at an impressed voltage of 5 kV/mm and a dielectric loss of 1.0% or less, along with the rate of change of the electrostatic capacitance against temperature changes satisfying the B-level characteristic standard stipulated in the JIS Standard in the temperature range of -25 °C to +85 °C and X7R-level characteristic standard stipulated in the EIA standard in the temperature range of -55 °C to +125 °C.

When the capacitor is used under a high electric field strength of 10 kV/mm, the insulation resistance represented by the product CR at 25 °C and 150 °C show a high value of 4900  $\Omega \cdot \text{F}$  or more and 200  $\Omega \cdot \text{F}$  or more, respectively. The insulation breakdown voltage also shows high values of 12 kV/mm or more under the AC voltage and 14 kV/mm under the DC voltage. The mean life span in the acceleration test at 150 °C and DC 25 kV/mm is as long as 800 hours along with being free from rejections in the humidity resistance load test and being possible to sinter at a relatively low sintering temperature of 1300 °C.

On the contrary, when the  $\text{SiO}_2\text{-TiO}_2\text{-XO}$  oxides is outside of the composition range described above as in the samples No. 213 to 219 in TABLE 8, the sintering becomes insufficient or many samples are rejected in the humidity resistance load test even after sintering as seen in the samples No. 513 to 519 in TABLE 15.

When  $\text{Al}_2\text{O}_3$  and/or  $\text{ZrO}_2$  is allowed to contain in the oxides represented by  $\text{SiO}_2\text{-TiO}_2\text{-XO}$  as in the samples No. 211 and 212 in TABLE 8, a monolithic capacitor having the insulation resistances of 5300  $\Omega \cdot \text{F}$  or more and 300  $\Omega \cdot \text{F}$  or more at 25 °C and 150 °C, respectively, under an electric field strength of 10 kV/mm as the samples No. 511 and 512 in TABLE 15 can be obtained. However, when the amounts of addition of  $\text{Al}_2\text{O}_3$  and  $\text{ZrO}_2$  exceed 15 parts by weight and 5 parts by weight, respectively, as in the samples No. 217 and 218 in TABLE 8, the sintering property is extremely deteriorated as in the samples No. 217 and 218 in TABLE 8.

#### (Example 7)

After preparing and weighing  $\text{TiCl}_4$  and  $\text{Ba}(\text{NO}_3)_2$  having a variety of purity as starting materials, the compounds were precipitated as titanyl barium oxalate ( $\text{BaTiO}(\text{C}_2\text{O}_4) \cdot 4\text{H}_2\text{O}$ ) by adding oxalic acid. This precipitate was decomposed by heating at a temperature of 1000 °C or more to synthesize four kinds of barium titanate listed in TABLE 1.

Oxides, carbonates or hydroxides as each component of the first side component were weighed so as to be a composition ratio (mole ratio) of  $0.25\text{Li}_2\text{O}-0.65(0.30\text{TiO}_2 + 0.70\text{SiO}_2)-0.10\text{Al}_2\text{O}_3$  to obtain a powder by crushing and mixing.

Likewise, oxides, carbonates or hydroxides as each component of the second side component were weighed so as to be a composition ratio (mole ratio) of  $0.66\text{SiO}_2-0.17\text{TiO}_2-0.15\text{BaO}-0.02\text{MnO}$  to obtain a powder by crushing and mixing.

Oxide powders of the first and second side components were placed in separate platinum crucibles, respectively, and heated at 1500 °C. After quenching and crushing the mixture, each oxide powder with a mean particle size of 1  $\mu\text{m}$  or less was obtained.

In the next step,  $\text{BaCO}_3$  for adjusting the mole ratio Ba/Ti (m) in barium titanate,  $\text{Sc}_2\text{O}_3$ ,  $\text{Y}_2\text{O}_3$ ,  $\text{BaZrO}_3$ ,  $\text{MgO}$  and  $\text{MnO}$ , each having a purity of 99% or more, were prepared. These raw material powders and the oxides described above to be either the first or second side component were weighted so as to form the compositions shown in TABLE 1002 and TABLE 1003. The amounts of addition of the first and second side components are indicated by parts by weight relative to 100 parts by weight of the essential component represented by  $((\text{BaO})_m\text{TiO}_2 + \alpha\text{M}_2\text{O}_3 + \beta\text{BaZrO}_3 + \gamma\text{MgO} + \text{gMnO})$ .

\* indicates "out of the scope of the present invention"

Table 1002

Sample No.	Kind of BaTiO <sub>3</sub>	(BaO) <sub>m</sub> TiO <sub>2</sub> + αM <sub>2</sub> O <sub>3</sub> + βBaZrO <sub>3</sub> + γMgO + gMnO										Amount of addition of the first side component (parts by weight)	Amount of addition of the second side component (parts by weight)
		α		Total of α	β	γ	g	γ + g	m				
		Sc <sub>2</sub> O <sub>3</sub>	Y <sub>2</sub> O <sub>3</sub>										
*1001	1A	0	0.0008	0.0008	0.02	0.002	0.002	0.004	1.005	1	0		
*1002	1A	0.01	0.06	0.07	0.03	0.07	0.05	0.12	1.005	1	0		
*1003	1A	0.01	0.02	0.03	0	0.03	0.02	0.05	1.01	1.5	0		
*1004	1A	0.02	0.01	0.03	0.08	0.02	0.04	0.06	1.01	1.5	0		
*1005	1A	0.01	0.01	0.02	0.02	0.001	0.034	0.035	1.01	1	0		
*1006	1A	0.01	0.02	0.03	0.02	0.125	0.005	0.13	1.01	1	0		
*1007	1A	0.02	0.02	0.04	0.02	0.079	0.001	0.08	1.01	1	0		
*1008	1A	0.01	0.02	0.03	0.02	0.005	0.125	0.13	1.01	1	0		
*1009	1A	0.01	0.02	0.03	0.03	0.08	0.06	0.14	1.01	1	0		
*1010	1A	0.02	0.01	0.03	0.03	0.02	0.03	0.05	0.99	1	0		
*1011	1A	0.01	0.01	0.02	0.04	0.03	0.02	0.05	1.00	1	0		
*1012	1A	0.02	0	0.02	0.04	0.02	0.01	0.03	1.038	1	0		
*1013	1A	0.01	0.01	0.02	0.02	0.03	0.01	0.04	1.05	0	1		
*1014	1A	0.01	0.01	0.02	0.02	0.01	0.02	0.03	1.01	0	0		
*1015	1A	0.02	0.01	0.03	0.02	0.03	0.02	0.05	1.01	5	0		
*1016	1A	0.01	0.01	0.02	0.02	0.01	0.03	0.04	1.01	0	0		
*1017	1A	0.005	0.025	0.03	0.02	0.03	0.02	0.05	1.01	0	4		
*1018	1D	0.01	0.02	0.03	0.03	0.03	0.03	0.06	1.01	2	0		
1019	1A	0	0.001	0.001	0.02	0.002	0.0015	0.003	1.015	1	0		
1020	1B	0.02	0	0.02	0.03	0.02	0.01	0.03	1.02	1	0		
1021	1C	0.005	0.05	0.055	0.03	0.12	0.002	0.122	1.03	1	0		
1022	1A	0	0.04	0.04	0.03	0.03	0.04	0.07	1.02	1	0		
1023	1A	0	0.06	0.06	0.03	0.01	0.12	0.13	1.01	0	1		

Table 1003

Sample No.	(BaO) <sub>m</sub> TiO <sub>2</sub> + αM <sub>2</sub> O <sub>3</sub> + βBaZrO <sub>3</sub> + γMgO + gMnO										Amount of addition of the first side component	Amount of addition of the second side component
	Kind of BaTiO <sub>3</sub>	α		Total of α	β	γ	g	γ + g	m			
		Sc <sub>2</sub> O <sub>3</sub>	Y <sub>2</sub> O <sub>3</sub>									
1024	1A	0	0.01	0.01	0.005	0.01	0.02	0.03	1.01	1	0	
1025	1A	0	0.02	0.02	0.04	0.01	0.03	0.04	1.01	1	0	
1026	1A	0	0.03	0.03	0.06	0.03	0.02	0.05	1.01	1	0	
1027	1A	0.01	0.04	0.05	0.03	0.06	0.05	0.11	1.01	2	0	
1028	1A	0	0.005	0.005	0.02	0.005	0.006	0.011	1.001	2	0	
1029	1A	0	0.04	0.04	0.03	0.05	0.02	0.07	1.01	0	2	
1030	1A	0	0.02	0.02	0.03	0.02	0.03	0.05	1.035	2	0	
1031	1A	0	0.02	0.02	0.03	0.03	0.01	0.04	1.015	0.2	0	
1032	1A	0	0.02	0.02	0.03	0.02	0.03	0.05	1.01	3	0	
1033	1A	0	0.02	0.02	0.03	0.02	0.015	0.035	1.01	0	0.2	
1034	1A	0	0.02	0.02	0.03	0.03	0.01	0.04	1.01	0	3	

Organic solvents such as polyvinyl butyral binder and ethanol were added to the weighed compounds and the mixture was mixed in a ball mill in a wet state to prepare a ceramic slurry. This ceramic slurry was formed into a sheet by a doctor blade method to obtain a rectangular shaped green sheet with a thickness of 35 μm, followed by printing an

electroconductive paste mainly composed of Ni on the ceramic green sheet to form an electroconductive paste layer for forming inner electrodes.

Then, a plurality of the ceramic green sheets on which the electroconductive layer is formed were laminated so that the sides where the electroconductive paste is projected out are alternately placed with each other, thus obtaining a monolithic body. This monolithic body was heated at 350 °C in a N<sub>2</sub> atmosphere and, after allowing the binder to decompose, the monolithic body was fired at the temperatures shown in TABLE 1004 and TABLE 1005 in a reducing atmosphere comprising H<sub>2</sub>-N<sub>2</sub>-H<sub>2</sub>O gases under an oxygen partial pressure of 10<sup>-9</sup> to 10<sup>-12</sup> MPa for two hours, thereby obtaining a ceramic sintered body.

The both side faces of the ceramic sintered body were coated with a silver paste containing B<sub>2</sub>O<sub>3</sub>-Li<sub>2</sub>O-SiO<sub>2</sub>-BaO glass frits and fired at a temperature of 600 °C in a N<sub>2</sub> atmosphere, thereby obtaining outer electrodes electrically connected to the inner electrodes.

The overall dimensions of the monolithic ceramic capacitor thus obtained were 5.0 mm in width, 5.7 mm in length and 2.4 mm in thickness while the thickness of the dielectric ceramic layer was 30 μm. Total number of the effective dielectric ceramic layers were 57, the area of the confronting electrode per one layer being  $8.2 \times 10^{-6} \text{ m}^2$ .

Electric characteristics of these monolithic ceramic capacitors were measured. The electrostatic capacitance (C) and dielectric loss (tan δ) were measured using an automatic bridge type measuring instrument at 1 kHz, 1 Vrms and 25 °C and the dielectric constant (ε) was calculated from the electrostatic capacitance. Next, the insulation resistance was measured using an insulation resistance tester at 25 °C and 150 °C by impressing direct current voltages of 315 V (or 10 kV/mm) and 945 V (or 30 kV/mm) for 2 minutes, obtaining a product of the electrostatic capacitance and insulation resistance, or a product CR.

The rate of change of the electrostatic capacitance against temperature changes was also measured. The rate of change at -25 °C and 85 °C by taking the electrostatic capacitance at 20 °C as a standard ( $\Delta C/C_{20}$ ), the rate of change at -55 °C and 125 °C by taking the electrostatic capacitance at 20 °C as a standard ( $\Delta C/C_{25}$ ) and the maximum value of the rate of change ( $|\Delta C| \text{ max}$ ) as an absolute value were measured as the electrostatic capacitances against temperature changes.

The DC bias characteristic was also evaluated. First, the electrostatic capacitance when an AC voltage of 1 kHz and 1 Vrms was impressed was measured. Then, the electrostatic capacitance when a DC voltage of 150 V and an AC voltage of 1 kHz and 1 Vrms were simultaneously impressed was measured, thereby the rate of reduction of the electrostatic capacitance ( $\Delta C/C$ ) due to loading the DC voltage was calculated.

In the high temperature load test, a direct current voltage of 750 V (or 25 kV/mm) was impressed at 150 °C on 36 pieces of each sample to measure the time dependent changes of the insulation resistance. The time when the insulation resistance of each sample was reduced below 10<sup>6</sup> Ω was defined to be a life span time and mean life span time was evaluated.

In the humidity resistance test, the number of the test pieces having an insulation resistance of 10<sup>6</sup> Ω or less among the 72 test pieces were counted after impressing a DC voltage of 315 V under an atmospheric pressure of 2 atm (relative humidity 100%) at 120 °C for 250 hours.

Insulation breakdown voltages under AC and DC voltages were measured by impressing AC and DC voltages at a voltage increase rate of 100 V/sec.

The results described above are listed in TABLE 1004 and TABLE 1005.

Table 1004 \* indicates "out of the scope of the present invention"

Sample No.	Baking temp. (°C)	Dielectric constant	Dielectric loss $\tan \delta$ (%)	Ratio of temperature dependent capacitance change (%)				DC bias characteristic (%) $\Delta C/C$ 5kV/mm	Product CR ( $\Omega F$ )				Insulation breakdown voltage (kV/mm)		Humidity resistance test: Number of reject	Mean life span (h)	
				$\Delta C/C_{25}$					315V Impressed Voltage	945V Impressed Voltage	315V Impressed Voltage	945V Impressed Voltage	AC	DC			
				$\Delta C/C_{25}$	-25°C	-55°C	125°C										Maximum value
*1001	1300	1140	0.8	5.5	-12.5	4.6	-17.2	20	-17	5130	4870	210	200	12	14	0/72	890
*1002	1300	850	0.7	2.1	-7.2	4.4	-6.5	8	-12	8490	8070	190	180	12	14	0/72	850
*1003	1280	1020	0.6	2	-8	4.2	-8.2	9.1	-15	3140	2200	150	110	12	14	0/72	840
*1004	1300	1360	0.7	2.4	-14.7	5.1	-29.5	35.4	-26	5170	4910	230	220	12	14	0/72	150
*1005	1280	1220	0.7	3.2	-14.2	4.1	-17.9	23	-21	3070	2920	130	120	13	14	0/72	830
*1006	1360	1400	2.7	2.5	-8.3	4.3	-8.6	9	-35	5140	4880	220	210	12	14	61/72	180
*1007	Unmeasurable due to semiconductor formation																
*1008	1280	1340	0.8	3.5	-8.9	4.8	-18.6	25.1	-28	3140	2980	140	130	12	14	0/72	190
*1009	1280	880	2.6	3.5	-9.2	3.8	-9.1	9.9	-12	5010	4760	240	230	12	14	15/72	130
*1010	Unmeasurable due to semiconductor formation																
*1011	1300	1280	0.7	3.9	-9	4.7	-9.3	10.2	-25	3300	3140	160	150	10	12	0/72	110
*1012	Unmeasurable due to insufficient sintering																
*1013	Unmeasurable due to insufficient sintering																
*1014	Unmeasurable due to insufficient sintering																
*1015	1300	1280	2.3	4	-8.9	4.2	-9.4	10.9	-25	3310	3150	160	150	11	11	0/72	150
*1016	Unmeasurable due to insufficient sintering																
*1017	1300	1260	2.5	2.8	-8.4	3.1	-7.6	8.4	-23	3150	2990	180	170	10	11	0/72	170
*1018	1300	1030	0.7	2.5	-7.9	5.6	-9.4	9.2	-15	5120	4860	260	250	12	14	0/72	870
1019	1280	1580	0.7	2.5	-7.2	3.8	-8.5	8.8	-45	5070	4820	260	250	12	15	0/72	950
1020	1280	1520	0.6	2.6	-7	4	-8	8.6	-45	5050	4800	240	230	12	14	0/72	960
1021	1280	1080	0.6	3	-8	4.1	-8.2	9	-15	5290	5030	280	270	12	15	0/72	820
1022	1300	1320	0.6	2.5	-8.3	3.8	-8.6	8.8	-26	5160	4900	250	240	12	14	0/72	830
1023	1300	1050	0.7	2.4	-8.1	4.2	-8.4	8.9	-15	5230	4970	220	210	13	15	0/72	840



Table 1005

Sam- ple No.	Bak- ing temp. (°C)	Dielec- tric con- stant	Dielec- tric loss tan δ (%)	Ratio of temperature dependent capacitance change (%)				DC vias charac- teristic (%) Δ C/C 5kV/mm	Product CR (Ω·F)				Insula- tion break- down voltage (kV/mm)		Humidi- ty resis- tance load test: Num- ber of rejects	Mean life span (h)	
				Δ C/C <sub>20</sub>		Δ C/C <sub>25</sub>			315V Im- pressed Voltage	945V Im- pressed Voltage	315V Im- pressed Voltage	945V Im- pressed Voltage	AC	DC			
				-25°C	85°C	-55°C	125°C										
																	Maxi- mum value
1024	1300	1570	0.6	2.9	-7.9	4.1	-8.2	8.8	-45	5180	4920	230	220	12	14	0/72	890
1025	1300	1480	0.7	3.1	-7.1	4.3	-7.9	8.5	-40	5300	5040	270	260	12	15	0/72	900
1026	1280	1420	0.6	3.2	-8.1	3.8	-8.3	9	-35	5240	4980	280	270	13	14	0/72	830
1027	1280	1230	0.7	2.4	-7.5	3.9	-8.1	8.6	-22	5260	5000	220	210	12	14	0/72	840
1028	1300	1590	0.6	2.8	-7.6	4.2	-7.9	8.4	-45	5280	5020	200	190	12	14	0/72	860
1029	1300	1330	0.6	2.7	-7.2	4.3	-8.2	8.6	-26	5040	4790	210	200	12	14	0/72	950
1030	1300	1540	0.6	2.6	-7.9	4.5	-8.1	8.8	-42	5230	4970	240	230	13	14	0/72	900
1031	1280	1550	0.6	2.5	-8	4.1	-8.5	8.9	-42	5080	4830	270	260	12	15	0/72	870
1032	1280	1550	0.6	2	-8.1	3.9	-8.2	8.7	-42	5190	4930	210	200	12	14	0/72	890
1033	1300	1540	0.7	2.4	-7.8	3.9	-8.5	9.1	-42	5140	4880	210	200	12	14	0/72	860
1034	1300	1520	0.6	2.5	-7.6	4.2	-8.4	9.2	-43	5080	4830	230	220	12	14	0/72	840

It is evident from Table 1002 to TABLE 1005 that the monolithic ceramic capacitor according to the present invention has a capacitance decreasing ratio of as small as within -45% at an impressed voltage of 5 kV/mm and a dielectric loss of less than 1.0 %, wherein the rate of change against temperature changes satisfies both the B-level characteristic

standard stipulated in the JIS Standard in the temperature range of -25 °C to +85 °C and X7R-level characteristic standard stipulated in the EIA standard in the temperature range of -55 °C to +125 °C.

Moreover, the insulation resistances at 25 °C and 150 °C as expressed by the product CR show as high values as 5000  $\Omega \cdot F$  or more and 200  $\Omega \cdot F$  or more, respectively, when the ceramic capacitor is used under a high electric field strength of 10 kV/mm. The insulation breakdown voltage also shows high values of 12 kV/mm or more under the AC voltage and 14 kV/mm or more under the DC voltage. In addition, an acceleration test at 150 °C and DC 25 kV/mm gave a mean life span as long as 800 hours or more besides enabling a relatively low firing temperature of 1300 °C or less.

The reason why the composition was limited in the present invention will be described hereinafter.

In the composition of  $(\text{BaO})_m\text{TiO}_2 + \alpha\text{M}_2\text{O}_3 + \beta\text{BaZrO}_3 + \gamma\text{MgO} + g\text{MnO}$  (wherein  $\text{M}_2\text{O}_3$  represents at least one of either  $\text{Sc}_2\text{O}_3$  or  $\text{Y}_2\text{O}_3$ ,  $\alpha$ ,  $\beta$ ,  $\gamma$ , and  $g$  representing mole ratio, respectively), the  $\text{M}_2\text{O}_3$  content  $\alpha$  of less than 0.001 as shown in the sample No. 1001 is not preferable because the temperature characteristic does not satisfy the B-level characteristic / X7R characteristic. On the other hand, the  $\text{M}_2\text{O}_3$  content  $\alpha$  of more than 0.06 as shown in the sample No. 1002 is also not preferable because the specific dielectric constant is reduced to less than 1000. Accordingly, the preferable range of the  $\text{M}_2\text{O}_3$  content  $\alpha$  is  $0.001 \leq \alpha \leq 0.06$ .

It is not preferable that the  $\text{BaZrO}_3$  content  $\beta$  of zero as in the sample No. 1003 is not preferable since the insulation resistance is low and the voltage dependency of the insulating resistance is larger than that of the composition system containing  $\text{BaZrO}_3$ . It is also not preferable that the  $\text{BaZrO}_3$  content  $\beta$  is more than 0.06 as in the sample No. 1004 because the temperature characteristic does not satisfy the B-level characteristic / X7R characteristic, along with shortening

It is not preferable that the amount of addition of the first or second side component is zero as in the samples No. 1014 and 1016 because measurements are impossible due to insufficient sintering. It is not preferable that the amount of addition of the first or second side component exceeds 3.0 parts by weight as seen in the samples No. 1015 and 1017, because the dielectric loss exceeds 1.0% and the insulation resistance and insulation breakdown voltage are lowered along with shortening the mean life span. Accordingly, the preferable content of either the first or the second components is 0.2 to 3.0 parts by weight.

The contents of the alkali earth metal oxides contained in barium titanate as impurities are suppressed below 0.02% by weight because, when the contents of the alkali earth metal oxides exceeds 0.02% by weight as in the sample No. 1018, the dielectric constant is decreased.

(Example 8)

A material with a composition of  $\text{BaO}_{1.010} \cdot \text{TiO}_2 + 0.03\text{Y}_2\text{O}_3 + 0.02\text{BaZrO}_3 + 0.05\text{MgO} + 0.01 \text{ MnO}$  (mole ratio) was prepared using barium titanate in TABLE 1A as a dielectric powder. A monolithic ceramic capacitor was produced by the same method as in Example 1, except that an oxide represented by  $\text{Li}_2\text{O} \cdot (\text{Si}, \text{Ti})\text{O}_2 \cdot \text{MO}$  shown in Table 1006, having a mean particle size of 1  $\mu\text{m}$  or less produced by heating the material described above at 1200 to 1500 °C, was added as the first side component. The overall dimensions of the monolithic ceramic capacitor produced is the same as in Example 1.

Table 1006

Sample No.	The first side component					
	Amount of addition (parts by weight)	Composition (mol %, except w)				
		Li <sub>2</sub> O	(Si <sub>w</sub> Ti <sub>1-w</sub> )O <sub>2</sub>	w	Al <sub>2</sub> O <sub>3</sub>	ZrO <sub>2</sub>
1101	1	20	80	0.3	0	0
1102	1	10	80	0.6	5	5
1103	0.8	10	70	0.5	20	0
1104	0.8	35	45	1	10	10
1105	1.5	45	45	0.5	10	0
1106	1.5	45	55	0.3	0	0
1107	1	20	70	0.6	5	5
1108	1	20	70	0.4	10	0
1109	1.2	30	60	0.7	5	5
1110	1.2	30	60	0.8	10	0
1111	2	40	50	0.6	5	5
1112	2	40	50	0.9	0	10
1113	1.5	10	85	0.4	5	0
1114	2	5	75	0.6	10	10
1115	1.2	20	55	0.5	25	0
1116	1	45	40	0.8	0	15
1117	0.8	50	45	0.7	5	0
1118	1.2	25	75	0.9	0	0
1119	1.5	25	75	1	0	0
1120	1	35	65	0.9	0	0
1121	1.5	35	65	1	0	0
1122	1.2	20	70	0.2	0	10

The electric characteristics were then measured by the same method as in Example 1. The results are shown in TABLE 1007.

Table 1007

Sample No.	Baking temp. (°C)	Dielectric constant	Dielectric loss tan δ (%)	Ratio of temperature dependent capacitance change (%)				DC vias charac- teristic (%)	Product CR (Ω·F)				Insulation break- down voltage (kV/mm)		Humidity resis- tant load test: Num- ber of reject	Mean life span (h)		
				Δ C/C <sub>20</sub>		Δ C/C <sub>25</sub>			Maxi- mum value		25°C		150°C				AC	DC
				-25°C	85°C	-55°C	125°C		315V Im- pressed Voltage	945V Im- pressed Voltage	315V Im- pressed Voltage	945V Im- pressed Voltage						
1101	1280	1430	0.8	2.5	-8.2	3.3	-8.4	9.4	-36	5120	4860	220	210	13	14	0/72	840	
1102	1280	1450	0.6	2.3	-8.4	3.8	-8.6	9.5	-37	5080	4830	230	220	13	14	0/72	890	
1103	1280	1490	0.6	2.4	-7.9	4.2	-8.2	9.9	-39	5160	4900	220	210	12	14	0/72	850	
1104	1300	1470	0.7	2.1	-7.8	4.6	-8.1	8.9	-39	5200	4940	210	200	12	15	0/72	830	
1105	1300	1430	0.6	2	-7.8	3.5	-8.3	9.4	-36	5180	4920	230	220	13	14	0/72	900	
1106	1280	1450	0.7	2.6	-8.2	3.7	-8.6	9.2	-37	5090	4840	210	200	12	14	0/72	870	
1107	1280	1430	0.8	2.8	-8.6	3.8	-8.9	9.3	-36	5070	4820	220	210	13	14	0/72	830	
1108	1280	1440	0.6	2.9	-8.4	4.1	-8.7	8.9	-36	5030	4780	220	210	12	14	0/72	890	
1109	1280	1480	0.7	2.4	-7.6	4.2	-8.2	8.8	-39	5220	4960	210	200	12	14	0/72	910	
1110	1300	1470	0.6	2.6	-7.4	3.8	-8.3	8.8	-39	5080	4830	230	220	13	14	0/72	870	
1111	1300	1420	0.8	2.4	-8	3.9	-8.1	9	-36	5100	4850	200	190	12	14	0/72	850	
1112	1280	1410	0.6	2.2	-8.4	3.9	-8.6	9.1	-35	5180	4920	210	200	12	14	0/72	840	
1113	1350	Unmeasurable due to insufficient sintering																
1114	1350	Unmeasurable due to insufficient sintering																
1115	1350	1460	1.5	2.8	-7.8	4.2	-8.2	8.9	-37	4820	4580	190	180	11	13	32/72	180	
1116	1350	Unmeasurable due to insufficient sintering																
1117	1350	Unmeasurable due to insufficient sintering																
1118	1300	1420	0.8	2.9	-7.9	4.8	-8.3	9.1	-36	5160	4900	210	200	12	14	0/72	860	
1119	1350	1430	1.4	2.7	-8.2	4.2	-8.4	9.2	-36	4890	4650	190	180	11	13	16/72	150	
1120	1300	1450	0.7	2.5	-8	4	-8.2	9.4	-37	5130	4870	210	200	12	14	0/72	840	
1121	1350	1460	1.4	2.2	-8.1	3.8	-8.6	9.1	-38	4950	4700	190	180	11	13	36/72	140	
1122	1350	1450	1.3	2.3	-8.3	3.8	-8.7	9.1	-38	4870	4630	190	180	11	13	25/72	190	

As is evident from TABLE 1006 and TABLE 1007, preferable results are obtained in the samples No. 1101 to 1112, 1118 and 1120, in which oxides with compositions within or on the boundary lines of the area surrounded by the straight lines connecting each spot indicated by A (X = 20, y = 80, z = 0), B (X = 10, y = 80, z = 10), C (X = 10, y = 70, z = 20),

D ( $X = 35$ ,  $y = 45$ ,  $z = 20$ ), E ( $X = 45$ ,  $y = 45$ ,  $z = 10$ ) and F ( $x = 45$ ,  $y = 55$ ,  $z = 0$ ), (wherein  $x$ ,  $y$  and  $z$  represent mole % and  $w$  represents mole ratio, which is in the range of  $0.3 \leq w < 1.0$  in the composition on the line A - F) of the three component phase diagram of the oxides represented by  $\text{Li}_2\text{O} \cdot (\text{Si}_w, \text{Ti}_{1-w})\text{O}_2$ -MO shown in FIG. 4 are added, wherein the samples have a capacitance decreasing ratio of as small as within -45% at an impressed voltage of 5 kV/mm and a dielectric loss of 1.0% or less, along with the rate of change of the electrostatic capacitance against temperature changes satisfying the B-level characteristic standard stipulated in the JIS Standard in the temperature range of -25 °C to +85 °C and X7R-level characteristic standard stipulated in the EIA standard in the temperature range of -55 °C to +125 °C.

Moreover, the insulation resistances at 25 °C and 150 °C as expressed by the product CR show as high values as 5000  $\Omega \cdot \text{F}$  or more and 200  $\Omega \cdot \text{F}$  or more, respectively, when the ceramic capacitor is used under a high electric field strength of 10 kV/mm. The insulation breakdown voltage also shows high values of 12 kV/mm or more under the AC voltage and 14 kV/mm or more under the DC voltage. In addition, an acceleration test at 150 °C and DC 25 kV/mm gave a mean life span as long as 800 hours or more besides enabling a relatively low firing temperature of 1300 °C or less.

On the contrary, when the  $\text{Li}_2\text{O} \cdot (\text{Si}_w, \text{Ti}_{1-w})\text{O}_2$ -MO oxides is outside of the composition range described above as shown in the samples No. 1113 to 1117, and 1119, the sintering becomes insufficient or many samples are rejected in the humidity resistance load test even after sintering. The samples with the composition falling on the line A-F and  $W = 1.0$  as in the samples No. 1119 and 1121 have high sintering temperature along with causing many rejects in the humidity resistance load test. When the value of  $w$  is less than 0.30 as shown in the sample No. 1122, the sintering temperature becomes high along with causing many rejects in the humidity resistance load test.

#### (Example 9)

A material with a composition of  $\text{BaO}_{1.010} \cdot \text{TiO}_2 + 0.03\text{Sc}_2\text{O}_3 + 0.015\text{BaZrO}_3 + 0.05\text{MgO} + 0.01 \text{ MnO}$  (mole ratio) was prepared using barium titanate in TABLE 1A as a dielectric powder. A monolithic ceramic capacitor was produced by the same method as in Example 1, except that an oxide  $\text{Li}_2\text{O} \cdot \text{TiO}_2 \cdot \text{XO}$  shown in Table 1008, having a mean particle size of 1  $\mu\text{m}$  or less produced by heating the material described above at 1200 to 1500 °C, was added as the second side component. The amounts of addition of  $\text{Al}_2\text{O}_3$  and  $\text{ZrO}_2$  correspond to parts by weight relative to 100 parts by weight of  $(x\text{SiO}_2 - y\text{TiO}_2 - z\text{XO})$ . The overall dimensions of the monolithic ceramic capacitor produced is the same as in Example 1.

Table 1008

Sample No.	The second side component														
	Amount of addition	Essential component (mol %)										Added component (parts by weight)			
		XO													
		SiO <sub>2</sub>	TiO <sub>2</sub>	BaO	CaO	SrO	MgO	ZnO	MnO	Total					
1201	1	85	1	1	0	0	0	4	9	14	0	0			
1202	1	35	51	0	10	0	0	0	4	14	0	0			
1203	1	30	20	0	30	0	15	4	1	50	0	0			
1204	1	39	1	20	20	2	0	13	5	60	0	0			
1205	1	70	10	5	5	0	0	10	0	20	0	0			
1206	1	45	10	0	0	0	0	15	30	45	0	0			
1207	1	50	20	10	10	3	7	0	0	30	0	0			
1208	1	50	30	0	16	0	0	0	4	20	0	0			
1209	1	35	30	25	10	0	0	0	0	35	0	0			
1210	1	40	40	10	0	0	0	5	5	20	0	0			
1211	1	45	22	3	30	0	0	0	0	33	15	0			
1212	1	45	22	3	30	0	0	0	0	33	10	5			
1213	1	65	25	5	5	0	0	0	0	10	0	0			
1214	1	25	40	15	0	10	0	5	5	35	0	0			
1215	1	30	10	30	25	0	0	5	0	60	0	0			
1216	1	50	0	35	15	0	0	0	0	50	0	0			
1217	1	45	22	30	0	0	3	0	0	33	25	0			
1218	1	45	22	30	0	3	0	0	0	33	0	15			
1219	1	30	60	10	0	0	0	0	0	10	0	0			

The electric characteristics were then measured by the same method as in Example 1. The results are shown in TABLE 1009.

Table 1009

Sample No.	Baking temp. (°C)	Dielectric constant	Dielectric loss tan δ (%)	Ratio of temperature dependent capacitance change (%)					DC vias characteristic (%)	Product CR (Ω.F)				Insulation breakdown voltage (kV/mm)		Humidity resistance load test. Number of reject	Mean life span (h)
				Δ C/C <sub>25</sub>						25°C				150°C			
				Δ C/C <sub>20</sub>						315V Im-pressed Voltage				945V Im-pressed Voltage			
				-25°C	85°C	-55°C	125°C	Maximum value						AC	DC		
1201	1300	1520	1.3	2.4	-7.2	2.5	-7.5	8.1	-41	5010	4760	200	190	12	14	0/72	820
1202	1300	1530	1.4	2.3	-7.5	2.4	-7.8	8.2	-41	5000	4750	210	200	12	14	0/72	810
1203	1300	1550	1.3	2.2	-7.8	2.3	-7.9	8	-42	5020	4770	200	190	13	14	0/72	800
1204	1300	1540	1.3	2.3	-7.7	2.4	-7.8	8.3	-41	5010	4760	200	190	12	15	0/72	860
1205	1300	1520	1.3	2.4	-7.5	2.2	-7.6	8.1	-41	5030	4780	200	190	12	14	0/72	830
1206	1300	1530	1.3	2.2	-7.4	2	-7.8	8.1	-41	5010	4760	200	190	13	15	0/72	890
1207	1300	1560	1.3	2.3	-7.6	2.1	-7.9	8.2	-42	5000	4750	210	200	12	14	0/72	870
1208	1300	1520	1.3	2.4	-7.5	2.3	-8	8.2	-41	5010	4760	200	190	12	14	0/72	850
1209	1300	1530	1.3	2.2	-7.1	2.4	-7.8	8.3	-41	5020	4770	200	190	13	14	0/72	820
1210	1300	1510	1.4	2.3	-7.5	2.4	-7.9	8.1	-41	5030	4780	200	190	12	14	0/72	810
1211	1300	1530	1.3	2.5	-7.2	2.5	-7.8	8	-41	5420	5150	320	300	12	14	0/72	860
1212	1300	1540	1.3	2.1	-7.1	2.2	-7.6	8.1	-41	5410	5140	300	290	12	14	0/72	840
1213	1350	1550	1.5	2	-7	2.3	-7.6	8.2	-42	4830	4590	200	190	11	13	52/72	160
1214	1350								Unmeasurable due to insufficient sintering								
1215	1350								Unmeasurable due to insufficient sintering								
1216	1350	1560	1.6	2.3	-7.2	2.4	-7.8	8.2	-42	4790	4550	200	190	11	13	72/72	180
1217	1350								Unmeasurable due to insufficient sintering								
1218	1350								Unmeasurable due to insufficient sintering								
1219	1350								Unmeasurable due to insufficient sintering								

As is evident from TABLE 1008 and TABLE 1009, preferable results are obtained in the samples No. 1201 to 1212 in which oxides with compositions within or on the boundary lines of the area surrounded by the straight lines connecting each spot indicated by A (X = 85, y = 1, z = 14), B (X = 35, y = 51, z = 14), C (X = 30, y = 20, z = 50) and D (X = 39,

y = 1, z = 60), (wherein x, y and z represent mole %) of the three component phase diagram of the oxides represented by  $\text{SiO}_2\text{-TiO}_2\text{-XO}$  shown in FIG. 5 are added, wherein the samples have a capacitance decreasing ratio of as small as -45% at an impressed voltage of 5 kV/mm and a dielectric loss of 1.0% or less, along with the rate of change of the electrostatic capacitance against temperature changes satisfying the B-level characteristic standard stipulated in the JIS Standard in the temperature range of -25 °C to +85 °C and X7R-level characteristic standard stipulated in the EIA standard in the temperature range of -55 °C to +125 °C.

Moreover, the insulation resistances at 25 °C and 150 °C as expressed by the product CR show as high values as 5000  $\Omega \cdot \text{F}$  or more and 200  $\Omega \cdot \text{F}$  or more, respectively, when the ceramic capacitor is used under a high electric field strength of 10 kV/mm. The insulation breakdown voltage also shows high values of 12 kV/mm or more under the AC voltage and 14 kV/mm or more under the DC voltage. In addition, an acceleration test at 150 °C and DC 25 kV/mm gave a mean life span as long as 800 hours or more besides enabling a relatively low firing temperature of 1300 °C or less.

When the composition of the oxide represented by  $\text{SiO}_2\text{-TiO}_2\text{-XO}$  is out of the composition range described above, on the other hands, sintering becomes insufficient as seen in the samples No. 1213 to 1219 or many rejects occur in the humidity resistance load test even after the sintering.

When  $\text{Al}_2\text{O}_3$  and/or  $\text{ZrO}_2$  is allowed to contain in the oxides represented by  $\text{SiO}_2\text{-TiO}_2\text{-XO}$  as in the samples No. 1211 and 1212, a monolithic capacitor having the insulation resistances of 5400  $\Omega \cdot \text{F}$  or more and 300  $\Omega \cdot \text{F}$  or more at 25 °C and 150 °C, respectively, under an electric field strength of 10 kV/mm can be obtained. However, when the amounts of addition of  $\text{Al}_2\text{O}_3$  and  $\text{ZrO}_2$  exceed 15 parts by weight and 5 parts by weight, respectively, the sintering property is extremely deteriorated as in the samples No. 1217 and 1218.

(Example 10)

Four kinds of barium titanate ( $\text{BaTiO}_3$ ) in TABLE 1, an oxide powder as a first side component and an oxide powder as a second side component were obtained by the same method as in Example 1.

Then,  $\text{BaCO}_3$  for adjusting the Ba/Ti mole ratio m in barium titanate, and  $\text{Sc}_2\text{O}_3$ ,  $\text{Y}_2\text{O}_3$ ,  $\text{BaZrO}_3$  and MnO with purity of 99% or more were prepared. These raw material powder and oxide powder to be either the first or second side component were weighed so as to be the composition shown in TABLE 1010 and TABLE 1011. The amount of addition of the first or second side component corresponds to the amount relative to 100 parts of the essential component of  $((\text{BaO})_m\text{TiO}_2 + \alpha\text{M}_2\text{O}_3 + \beta\text{BaZrO}_3 + \gamma\text{MnO})$ . A monolithic ceramic capacitor was produced using this weighed compounds by the same method as in Example 1. The overall dimensions of the monolithic ceramic capacitor produced are the same as in Example 1.



Table 1010

\*indicates "out of the scope of the present invention"

Sample No.	(BaO) <sub>m</sub> TiO <sub>2</sub> + αM <sub>2</sub> O <sub>3</sub> + βBaZrO <sub>3</sub> + γMnO										Amount of addition of the first side component (parts by weight)	Amount of addition of the second side component (parts by weight)
	Kind of BaTiO <sub>3</sub>	α			Total of α	β	γ	m				
		Sc <sub>2</sub> O <sub>3</sub>	Y <sub>2</sub> O <sub>3</sub>									
*1301	1A	0	0.0008	0.0008	0.0008	0.01	0.0015	1.005	1	0		
*1302	1A	0.03	0.045	0.075	0.075	0.02	0.15	1.01	1	0		
*1303	1A	0.01	0.01	0.02	0.02	0	0.042	1.01	1.5	0		
*1304	1A	0.02	0.01	0.03	0.03	0.07	0.06	1.01	1.5	0		
*1305	1A	0	0.02	0.02	0.02	0.02	0.001	1.01	1	0		
*1306	1A	0.02	0	0.02	0.02	0.03	0.14	1.01	1	0		
*1307	1A	0	0.03	0.03	0.03	0.03	0.06	0.99	1	0		
*1308	1A	0.005	0.015	0.02	0.02	0.04	0.041	1	1	0		
*1309	1A	0	0.02	0.02	0.02	0.03	0.04	1.038	1	0		
*1310	1A	0	0.03	0.03	0.03	0.02	0.06	1.045	0	1		
*1311	1A	0	0.02	0.02	0.02	0.02	0.04	1.01	0	0		
*1312	1A	0.01	0.02	0.03	0.03	0.02	0.06	1.01	5	0		
*1313	1A	0.02	0.01	0.03	0.03	0.02	0.063	1.02	0	0		
*1314	1A	0	0.02	0.02	0.02	0.03	0.04	1.01	0	4		
*1315	1D	0	0.03	0.03	0.03	0.03	0.06	1.02	1.5	0		

Table 1011

Sample No.	(BaO) <sub>m</sub> TiO <sub>2</sub> + αM <sub>2</sub> O <sub>3</sub> + βBaZrO <sub>3</sub> + γMnO							Amount of addition of the first side component (parts by weight)	Amount of addition of the second side component (parts by weight)
	Kind of BaTiO <sub>3</sub>	α		Total of α	β	γ	m		
		Sc <sub>2</sub> O <sub>3</sub>	Y <sub>2</sub> O <sub>3</sub>						
1316	A	0	0.001	0.001	0.02	0.002	1.01	1	0
1317	B	0	0.02	0.02	0.03	0.04	1.02	1	0
1318	C	0.01	0	0.01	0.03	0.13	1.03	1	0
1319	A	0.01	0.04	0.05	0.04	0.1	1.02	1	0
1320	A	0.02	0.04	0.06	0.03	0.12	1.01	0	1
1321	A	0	0.01	0.01	0.005	0.02	1.01	1	0
1322	A	0.01	0.01	0.02	0.04	0.04	1.01	2	0
1323	A	0	0.03	0.03	0.06	0.06	1.01	1	0
1324	A	0.02	0.02	0.04	0.03	0.078	1.01	2	0
1325	A	0	0.01	0.01	0.02	0.02	1.001	2	0
1326	A	0	0.005	0.005	0.04	0.01	1.02	0	2
1327	A	0.01	0.01	0.02	0.03	0.04	1.035	2	0
1328	A	0.02	0	0.02	0.03	0.04	1.025	0.2	0
1329	A	0.01	0.01	0.02	0.03	0.04	1.01	3	0
1330	A	0	0.02	0.02	0.03	0.041	1.01	0	0.2
1331	A	0	0.02	0.02	0.03	0.04	1.01	0	3

The electric characteristics were measured by the same method as in Example 1. The results are shown in TABLE 1012 and TABLE 1013.

Table 1012

\* indicates "out of the scope of the present invention"

Sam- ple No.	Bak- ing temp. (°C)	Dielec- tric con- stant	Dielec- tric loss tan δ (%)	Ratio of temperature dependent capacitance change (%)				DC vias charac- teristic (%) Δ C/C 5kV/mm	Product CR (Ω·F)				Insula- tion break- down voltage (kV/mm)		Humidi- ty resis- tance load test: Num- ber of reject	Mean life span (h)	
				Δ C/C <sub>25</sub>					315V Im- pressed Voltage	945V Im- pressed Voltage	315V Im- pressed Voltage	945V Im- pressed Voltage	150°C				
				-25°C	85°C	125°C	Maxi- mum value						25°C	AC			DC
*1301	1300	1570	0.7	5	-12	4.2	-16.7	16.7	-42	5100	4850	200	190	12	14	0/72	900
*1302	1300	820	0.7	2.2	-6.7	4	-6	6.7	-9	5500	5230	180	170	12	14	0/72	910
*1303	1300	1450	0.7	2.1	-7.5	4	-7.7	8.2	-36	3020	2110	100	70	12	14	0/72	890
*1304	1300	1350	0.7	2.3	-14.2	5.3	-28	28	-27	5100	4850	200	190	12	14	0/72	120
*1305	Unmeasurable due to semiconductor formation																
*1306	1280	1420	0.8	3.5	-8.4	4.5	-18.1	18	-36	3120	2960	140	130	12	14	0/72	140
*1307	Unmeasurable due to semiconductor formation																
*1308	1300	1410	0.7	3.5	-8.5	4.3	-8.8	9.1	-35	3100	2950	130	120	10	12	0/72	100
*1309	Unmeasurable due to insufficient sintering																
*1310	Unmeasurable due to insufficient sintering																
*1311	Unmeasurable due to insufficient sintering																
*1312	1300	1330	2.3	3.6	-8.4	4.1	-8.9	9.2	-27	3330	3160	140	130	11	11	0/72	130
*1313	Unmeasurable due to insufficient sintering																
*1314	1300	1450	2.5	2.7	-7.9	3.2	-7.1	7.9	-36	3150	2990	160	150	10	11	0/72	160
*1315	1300	1310	0.7	2.6	-7.4	5.5	-8.9	9.2	-25	5060	4810	230	220	12	14	0/72	920

Table 1013

Sam- ple No.	Bak- ing temp. (°C)	Dielec- tric con- stant	Dielec- tric loss tan δ (%)	Ratio of temperature dependent capacitance change (%)				DC vias charac- teristic (%) Δ C/C 5kV/mm	Product CR (Ω.F)				Insula- tion break- down voltage (kV/mm)		Humidi- ty resis- tance load test: Num- ber of reject	Mean life span (h)		
				Δ C/C <sub>25</sub>			Maxi- mum value		25°C				150°C					
				-25°C	85°C	-55°C			125°C	315V Im- pressed Voltage	945V Im- pressed Voltage	315V Im- pressed Voltage	945V Im- pressed Voltage	315V Im- pressed Voltage			945V Im- pressed Voltage	
1316	1280	1590	0.7	2.5	-6.7	3	-8	8.4	4900	4660	230	220	220	12	14	970		
1317	1280	1450	0.6	2.7	-6.5	3.8	-7.5	8	4960	4710	220	210	220	13	15	960		
1318	1280	1570	0.7	3	-7.5	3.5	-7.7	8.2	5000	4750	250	240	240	12	14	850		
1319	1300	1110	0.6	2.6	-7.8	3.7	-8.5	8.8	5100	4850	210	200	200	12	14	840		
1320	1300	940	0.6	2.5	-7.6	4.1	-7.8	8.1	5120	4860	190	180	180	12	14	900		
1321	1300	1530	0.6	2.7	-7.4	4	-7.6	8	5070	4820	200	190	190	12	14	890		
1322	1300	1460	0.7	3	-6.6	4.2	-7.4	7.9	5200	4940	250	240	240	13	15	910		
1323	1300	1340	0.6	3.1	-7.6	4	-7.8	8.2	5230	4970	250	240	240	13	15	870		
1324	1280	1230	0.7	2.5	-7	3.7	-7.6	7.9	5260	5000	210	200	200	12	14	860		
1325	1300	1560	0.6	2.7	-7.1	4	-7.3	7.7	5100	4850	190	180	180	12	14	890		
1326	1300	1580	0.6	2.6	-6.7	4.1	-7.5	7.8	5020	4770	190	180	180	12	14	950		
1327	1300	1430	0.7	2.5	-7.4	4.2	-7.5	7.8	5110	4860	230	220	220	13	15	930		
1328	1280	1430	0.6	2.5	-7.5	4.1	-7.8	8	4980	4730	250	240	240	12	14	900		
1329	1280	1440	0.6	2.3	-7.6	4	-7.7	8	5080	4830	190	180	180	12	14	890		
1330	1300	1430	0.7	2.7	-7.3	3.8	-7.8	8.1	5060	4810	200	190	190	13	15	910		
1331	1300	1340	0.6	2.6	-7.1	4	-7.9	8.2	4990	4740	200	190	190	12	14	880		

As evident from TABLE 1012 and TABLE 1013, the ceramic capacitor according to the present invention has a capacitance decreasing ratio of as small as within -40% at an impressed DC voltage of 5 kV/mm and a dielectric loss of 1.0% or less, along with the rate of change of the electrostatic capacitance against temperature changes satisfying

the B-level characteristic standard stipulated in the JIS Standard in the temperature range of -25 °C to +85 °C and X7R-level characteristic standard stipulated in the EIA standard in the temperature range of - 55 °C to +125 °C.

Moreover, the insulation resistances at 25 °C and 150 °C as expressed by the product CR show as high values as 4900  $\Omega \cdot F$  or more and 190  $\Omega \cdot F$  or more, respectively, when the ceramic capacitor is used under a high electric field strength of 10 kV/mm. The insulation breakdown voltage also shows high values of 12 kV/mm or more under the AC voltage and 14 kV/mm or more under the DC voltage. In addition, an acceleration test at 150 °C and DC 25 kV/mm gave a mean life span as long as 800 hours or more besides enabling a relatively low firing temperature of 1300 °C or less.

The reason why the composition was limited in the present invention will be described hereinafter.

In the composition of  $(\text{BaO})_m\text{TiO}_2 + \alpha\text{M}_2\text{O}_3 + \beta\text{BaZrO}_3 + \gamma\text{MnO}$  (wherein  $\text{M}_2\text{O}_3$  represents at least one of either  $\text{Sc}_2\text{O}_3$  and  $\text{Y}_2\text{O}_3$ ,  $\alpha$ ,  $\beta$ , and  $\gamma$  representing mole ratio, respectively), the  $\text{M}_2\text{O}_3$  content  $\alpha$  of less than 0.001 as shown in the sample No. 1301 is not preferable because the temperature characteristic does not satisfy the B-level characteristic / X7R characteristic. On the other hand, the  $\text{M}_2\text{O}_3$  content  $\alpha$  of more than 0.06 as shown in the sample No. 1302 is also not preferable because the specific dielectric constant is reduced to less than 900. Accordingly, the preferable range of the  $\text{M}_2\text{O}_3$  content  $\alpha$  is  $0.001 \leq \alpha \leq 0.06$ .

It is not preferable that the  $\text{BaZrO}_3$  content  $\beta$  of zero as in the sample No. 1303 is not preferable since the insulation resistance is low and the voltage dependency of the insulating resistance is larger than that of the composition system containing  $\text{BaZrO}_3$ . It is also not preferable that the  $\text{BaZrO}_3$  content  $\beta$  is more than 0.06 as in the sample No. 1304 because the temperature characteristic does not satisfy the B-level characteristic / X7R characteristic, along with shortening the mean life span. Accordingly, the preferable range of the  $\text{BaZrO}_3$  content  $\beta$  is  $0.005 \leq \beta \leq 0.06$ .

It is not preferable that, as seen in the sample No. 1305, the  $\text{MgO}$  content  $\gamma$  is 0.001 since measurements becomes impossible due to semiconductor formation. It is not preferable that the  $\text{MgO}$  content  $\gamma$  exceeds 0.13 as in the sample No. 1306, because the temperature characteristic does not satisfy the X7R characteristic besides the insulation resistance is low and mean life span is shortened. Accordingly, the preferable range of the  $\text{MgO}$  content  $\gamma$  is in the range of  $0.001 < \gamma \leq 0.13$ .

It is not preferable that the  $\text{BaO}/\text{TiO}_2$  ratio  $m$  is less than 1.000 as in the sample No. 1307 because measurements are impossible due to formation of semiconductors. It is also not preferable that, as seen in the sample No. 1308, the  $\text{BaO}/\text{TiO}_2$  ratio  $m$  is 1.000 since the insulation resistance as well as the AC and DC breakdown voltage becomes low along with shortening the mean life span. It is not preferable, on the other hand, that the  $\text{BaO}/\text{TiO}_2$  ratio  $m$  is over 1.035 as in the samples No. 1309 and 1310 since measurements becomes impossible due to insufficient sintering. Accordingly, the  $\text{BaO}/\text{TiO}_2$  ratio  $m$  in the range of  $1.000 < m \leq 1.035$  is preferable.

It is not preferable that the amount of addition of the first or second side component is zero as in the samples No. 1311 and 1313 because measurements are impossible due to insufficient sintering. It is not preferable that the amount of addition of the first or second side component exceeds 3.0 parts by weight as seen in the samples No. 1312 and 1314, on the other hand, because the dielectric loss exceeds 1.0% and the insulation resistance and insulation breakdown voltage are lowered along with shortening the mean life span. Accordingly, the preferable content of either the first or the second components is in the range of 0.2 to 3.0 parts by weight.

The contents of the alkali earth metal oxides contained in barium titanate as impurities are suppressed below 0.02% by weight because, as in the sample No. 1315, when the contents of the alkali earth metal oxides exceeds 0.02% by weight, the dielectric constant is decreased.

#### (Example 11)

A material with a composition of  $\text{BaO}_{1.010} \cdot \text{TiO}_2 + 0.02\text{Y}_2\text{O}_3 + 0.01\text{BaZrO}_3 + 0.04 \text{ MnO}$  (mole ratio) was prepared using barium titanate in TABLE 1A as a dielectric powder. A monolithic ceramic capacitor was produced by the same method as in Example 1, except that an oxide represented by  $\text{Li}_2\text{O}-(\text{Si,Ti})\text{O}_2\text{-MO}$  as shown in Table 1006, having a mean particle size of 1  $\mu\text{m}$  or less produced by heating the material described above at 1200 to 1500 °C, was added as the first side component. The overall dimensions of the monolithic ceramic capacitor produced is the same as in Example 1. The electric characteristics were then measured by the same method as in Example 1. The results are shown in TABLE 1014. The sample No. 1401 to 1422 in TABLE 1014 correspond to the samples NO. 1101 to 1122 in TABLE 1006, respectively. For example, the sample No. 1401 in TABLE 1014 was obtained by adding the side component of the sample No. 1101 in TABLE 1006.

Table 1014

Sample No.	Baking temp. (°C)	Dielectric constant	Dielectric loss tan δ (%)	Ratio of temperature dependent capacitance change (%)					DC bias characteristic (%) ΔC/C 5kV/mm	Product CR (Ω·F)				Insulation breakdown voltage (kV/mm)		Humidity resistance test: Number of reject	Mean life span (h)
				ΔC/C <sub>20</sub>						315V Im-pressed Voltage	945V Im-pressed Voltage	315V Im-pressed Voltage	945V Im-pressed Voltage				
				ΔC/C <sub>25</sub>													
				-25°C	85°C	-55°C	125°C	Maximum value									
1401	1280	1420	0.7	2.4	-8.1	3.2	-8.3	8.5	-35	4950	4700	210	200	12	14	0/72	900
1402	1300	1430	0.6	2.2	-8.3	3.7	-8.5	8.7	-37	4980	4730	220	210	13	15	0/72	910
1403	1280	1460	0.6	2.4	-7.8	4	-8.1	8.3	-36	5100	4850	200	190	12	14	0/72	860
1404	1300	1480	0.7	2.1	-7.9	4.1	-8	8.2	-37	5000	4750	230	220	12	15	0/72	870
1405	1300	1440	0.7	2.1	-7.8	3.7	-8.2	8.4	-36	5080	4830	220	210	13	14	0/72	880
1406	1300	1440	0.7	2.6	-8.1	3.6	-8.5	8.8	-36	5070	4820	200	190	13	15	0/72	910
1407	1300	1430	0.8	2.7	-8.5	3.7	-8.5	8.8	-35	5120	4860	210	200	13	14	0/72	930
1408	1280	1450	0.6	2.9	-8.4	4	-8.5	8.8	-37	5160	4900	210	200	12	14	0/72	970
1409	1280	1470	0.7	2.5	-7.9	4.1	-8.1	8.4	-36	5000	4750	200	190	12	14	0/72	900
1410	1280	1480	0.6	2.6	-7.6	3.7	-8.3	8.5	-37	5140	4880	220	210	13	14	0/72	880
1411	1300	1430	0.7	2.5	-7.5	3.8	-8.2	8.3	-35	5100	4850	210	200	13	15	0/72	890
1412	1280	1420	0.6	2.3	-8	3.7	-8.4	8.5	-35	5120	4860	200	190	12	14	0/72	930
1413	1350	Unmeasurable due to insufficient sintering															
1414	1350	Unmeasurable due to insufficient sintering															
1415	1350	1450	1.5	2.7	-7.9	4	-8	8.4	-37	4900	4660	180	170	11	13	45/72	140
1416	1350	Unmeasurable due to insufficient sintering															
1417	1350	Unmeasurable due to insufficient sintering															
1418	1300	1430	0.8	2.8	-8	4.2	-8.2	8.6	-35	5030	4780	200	190	11	14	0/72	900
1419	1350	1440	1.5	2.6	-8.1	4	-8.3	8.5	-36	4980	4730	190	180	11	13	23/72	130
1420	1300	1460	0.7	2.6	-8.3	3.9	-8	8.5	-37	5000	4750	200	190	12	13	0/72	880
1421	1350	1450	1.5	2.4	-8.2	3.7	-8.2	8.7	-37	4970	4720	190	180	11	13	39/72	150
1422	1350	1460	1.3	2.4	-8.3	3.7	-8.5	8.7	-37	4990	4740	190	180	11	13	36/72	160

As is evident from the samples NO. 1401 to 1412, 1418 and 1420 in TABLE 1014, preferable results are obtained in the samples in which oxides of the samples No. 1101 to 1112, 1118 and 1120 in TABLE 1006 with compositions

within or on the boundary lines of the area surrounded by the straight lines connecting each spot indicated by A ( $X = 20, y = 80, z = 0$ ), B ( $X = 10, y = 80, z = 10$ ), C ( $X = 10, y = 70, z = 20$ ), D ( $X = 35, y = 45, z = 20$ ), E ( $x = 45, y = 45, z = 10$ ) and F ( $x = 45, y = 55, z = 0$ ) (wherein  $x, y$  and  $z$  represent mole %,  $w$  represent mole ratio and  $w$  is in the range of  $0.3 \leq w < 1.0$  in the composition on the line A - F) inside of the three component phase diagram of the oxides represented by  $\text{Li}_2\text{O}-(\text{Si}_w, \text{Ti}_{1-w})\text{O}_2$ -MO shown in FIG. 4 are added as side components, wherein the samples have a capacitance decreasing ratio of as small as within -40% at an impressed voltage of 5 kV/mm and a dielectric loss of 1.0% or less, along with the rate of change of the electrostatic capacitance against temperature changes satisfying the B-level characteristic standard stipulated in the JIS Standard in the temperature range of  $-25^\circ\text{C}$  to  $+85^\circ\text{C}$  and X7R-level characteristic standard stipulated in the EIA standard in the temperature range of  $-55^\circ\text{C}$  to  $+125^\circ\text{C}$ .

Moreover, the insulation resistances at  $25^\circ\text{C}$  and  $150^\circ\text{C}$  as expressed by the product CR show as high values as  $4900 \Omega \cdot \text{F}$  or more and  $200 \Omega \cdot \text{F}$  or more, respectively, when the ceramic capacitor is used under a high electric field strength of 10 kV/mm. The insulation breakdown voltage also shows high values of 12 kV/mm or more under the AC voltage and 14 kV/mm or more under the DC voltage. In addition, an acceleration test at  $150^\circ\text{C}$  and DC 25 kV/mm gave a mean life span as long as 800 hours or more besides enabling a relatively low firing temperature of  $1300^\circ\text{C}$  or less.

On the contrary, when the oxides represented by  $\text{Li}_2\text{O}-(\text{Si}_w, \text{Ti}_{1-w})\text{O}_2$ -MO is outside of the composition range described above as shown in the samples No. 1113 to 1117, and 1119 in TABLE 1006, the sintering becomes insufficient or many samples are rejected in the humidity resistance load test even after sintering as seen in the samples NO. 1413 to 1417 and 1419 in TABLE 1014. The samples with the composition falling on the line A-F and  $W = 1.0$  as in the samples No. 1119 and 1121 in TABLE 1006 have high sintering temperature along with causing many rejects in the humidity resistance load test as seen in the samples No. 1419 and 1421 in TABLE 1014. When the value of  $w$  is less than 0.30 as shown in the samples No. 1122 in TABLE 1006, the sintering temperature becomes high along with causing many rejects in the humidity resistance load test as seen in the sample No. 1422 in TABLE 1014.

#### (Example 12)

A material with a composition of  $\text{BaO}_{1.010} \cdot \text{TiO}_2 + 0.02\text{Sc}_2\text{O}_3 + 0.01\text{BaZrO}_3 + 0.04 \text{MnO}$  (mole ratio) was prepared using barium titanate in TABLE 1A as a dielectric powder. A monolithic ceramic capacitor was produced by the same method as in Example 1, except that an oxide represented by  $\text{SiO}_2$ - $\text{TiO}_2$ -XO shown in Table 1008, having a mean particle size of  $1 \mu\text{m}$  or less produced by heating the material described above at  $1200$  to  $1500^\circ\text{C}$ , was added as the second side component. The overall dimensions of the monolithic ceramic capacitor produced is the same as in Example 1. The electric characteristics were then measured by the same method as in Example 1. The results are shown in TABLE 1015. In TABLE 1015, the samples NO. 1501 to 1519 corresponds to the samples NO. 1201 to 1219 in TABLE 1008. For example, the sample No. 1501 in TABLE 1015 was obtained by adding the side component of the sample No. 1201 in TABLE 1008.

Table 1015

Sam- ple No.	Bak- ing temp. (°C)	Dielec- tric con- stant	Dielec- tric loss tan δ (%)	Ratio of temperature dependent capacitance change (%)					DC vias charac- teristic (%) Δ C/C 5kV/mm	Product CR (Ω.F)				Insula- tion break- down voltage (kV/mm)		Humidi- ty resis- tance load test: Num- ber of rejects	Mean life span (h)	
				Δ C/C <sub>20</sub>						25°C								150°C
				-25°C	85°C	-55°C	Δ C/C <sub>25</sub>	125°C		Maxi- mum value	315V Im- pressed Voltage	945V Im- pressed Voltage	315V Im- pressed Voltage	945V Im- pressed Voltage	AC			DC
1501	1300	1440	1	2.3	-7.3	2.3	-7.6	8	-35	4950	4700	190	180	13	15	0/72	920	
1502	1300	1430	1.1	2.2	-7.2	2.2	-7.8	8.2	-35	4970	4720	200	190	12	14	0/72	890	
1503	1300	1460	1.2	2.1	-7.7	2.1	-7.7	8.1	-36	5000	4750	190	180	13	15	0/72	920	
1504	1300	1420	1.1	2.1	-7.6	2.3	-7.8	8.2	-34	5020	4770	230	220	13	15	0/72	960	
1505	1300	1470	1	2.3	-7.4	2.2	-7.6	8.1	-37	4960	4710	200	190	12	14	0/72	880	
1506	1300	1450	1.2	2.2	-7.2	2.1	-7.9	8.2	-36	5100	4850	200	190	12	15	0/72	940	
1507	1300	1420	1.2	2.3	-7.3	2.3	-7.9	8.2	-34	5020	4770	200	190	12	14	0/72	950	
1508	1300	1430	1.2	2.3	-7.5	2.4	-8.1	8.2	-35	5060	4810	230	220	12	14	0/72	870	
1509	1300	1440	1.1	2.1	-7.2	2.3	-7.8	8.2	-35	5050	4800	210	200	12	14	0/72	890	
1510	1300	1480	1.2	2.2	-7.3	2.4	-8	8.1	-37	5020	4770	200	190	12	14	0/72	900	
1511	1300	1460	1.1	2.3	-7.1	2.4	-7.8	8.3	-36	5290	5030	310	300	13	15	0/72	960	
1512	1300	1440	1.2	2.4	-7.3	2.3	-7.6	8.1	-35	5300	5040	320	300	12	14	0/72	950	
1513	1350	1420	1.1	2.3	-7.2	2	-7.7	8.1	-34	4820	4580	210	200	11	13	46/72	150	
1514	1350									Unmeasurable due to insufficient sintering								
1515	1350									Unmeasurable due to insufficient sintering								
1516	1350	1430	1.2	2.2	-7.3	2.5	-7.7	8.1	-34	4790	4550	210	200	11	13	72/72	130	
1517	1350									Unmeasurable due to insufficient sintering								
1518	1350									Unmeasurable due to insufficient sintering								
1519	1350									Unmeasurable due to insufficient sintering								

As is evident from Samples NO. 1501 to 1512 in TABLE 1015, preferable results are obtained in the samples, in which oxides of the samples NO. 1201 to 1212 in TABLE 1008 with compositions within or on the boundary lines of the



area surrounded by the straight lines connecting each spot indicated by A ( $X = 85, y = 1, z = 14$ ), B ( $X = 35, y = 51, z = 14$ ), C ( $X = 30, y = 20, z = 50$ ) and D ( $X = 39, y = 1, z = 60$ ), (wherein  $x, y$  and  $z$  represent mole %), of the three component phase diagram of the oxides represented by  $\text{SiO}_2\text{-TiO}_2\text{-XO}$  shown in FIG. 5 are added as a side component, wherein the samples have a capacitance decreasing ratio of as small as within -40% at an impressed voltage of 5 kV/mm and a dielectric loss of 1.0% or less, along with the rate of change of the electrostatic capacitance against temperature changes satisfying the B-level characteristic standard stipulated in the JIS Standard in the temperature range of -25 °C to +85 °C and X7R-level characteristic standard stipulated in the EIA standard in the temperature range of -55 °C to +125 °C.

Moreover, the insulation resistances at 25 °C and 150 °C as expressed by the product CR show as high values as 4900  $\Omega \cdot \text{F}$  or more and 190  $\Omega \cdot \text{F}$  or more, respectively, when the ceramic capacitor is used under a high electric field strength of 10 kV/mm. The insulation breakdown voltage also shows high values of 12 kV/mm or more under the AC voltage and 14 kV/mm or more under the DC voltage. In addition, an acceleration test at 150 °C and DC 25 kV/mm gave a mean life span as long as 800 hours or more and no rejections were found in the humidity resistance load test besides enabling a relatively low firing temperature of 1300 °C or less.

When the oxide  $\text{SiO}_2\text{-TiO}_2\text{-XO}$  has a composition outside of the composition described above as in the samples No. 1213 to 1219 in TABLE 1008, on the contrary, sintering becomes insufficient or many rejection appear in the humidity resistance load test even after sintering as seen in the samples No. 1513 to 1519 in TABLE 1015.

While a monolithic capacitor having an insulation resistance of 5290  $\Omega \cdot \text{F}$  or more and 310  $\Omega \cdot \text{F}$  or more at 25 °C and 150 °C, respectively, under a strong electric field of 10 kV/mm can be obtained as shown in the samples No. 1511 and 1512 in TABLE 1015 by allowing  $\text{Al}_2\text{O}_3$  and/or  $\text{ZrO}_2$  to contain in the  $\text{SiO}_2\text{-TiO}_2\text{-XO}$  oxides as in the sample No. 1211 and 1212 in Table 1008, sintering property is extremely decreased when  $\text{Al}_2\text{O}_3$  and  $\text{ZrO}_2$  are added in an amounts of 15 parts by weight or more and 5 parts by weight or more, respectively, as in the samples No. 1217 and 1218 in TABLE 1008.

#### (Example 13)

After preparing and weighing  $\text{TiCl}_4$  and  $\text{Ba}(\text{NO}_3)_2$  having a variety of purity as starting materials, the compounds were precipitated as titanyl barium oxalate ( $\text{BaTiO}(\text{C}_2\text{O}_4) \cdot 4\text{H}_2\text{O}$ ) by adding oxalic acid. This precipitate was decomposed by heating at a temperature of 1000 °C or more to synthesize four kinds of barium titanate listed in TABLE 1.

Oxides, carbonates or hydroxides as each component of the first side component were weighed so as to be a composition ratio (mole ratio) of  $0.25\text{Li}_2\text{O}-0.65(0.30\text{TiO}_2 + 0.70\text{SiO}_2)-0.10\text{Al}_2\text{O}_3$  to obtain a powder by crushing and mixing.

Likewise, oxides, carbonates or hydroxides as each component of the second side component were weighed so as to be a composition ratio (mole ratio) of  $0.66\text{SiO}_2-0.17\text{TiO}_2-0.15\text{BaO}-0.02\text{MnO}$  (mole ratio) to obtain a powder by crushing and mixing.

Oxide powders of the first and second side components were placed in separate platinum crucibles and heated at 1500 °C. After quenching and crushing the mixture, each oxide powder with a mean particle size of 1  $\mu\text{m}$  or less was obtained.

In the next step,  $\text{BaCO}_3$  for adjusting the mole ratio Ba/Ti (m) in barium titanate,  $\text{Eu}_2\text{O}_3$ ,  $\text{Gd}_2\text{O}_3$ ,  $\text{Tb}_2\text{O}_3$ ,  $\text{Dy}_2\text{O}_3$ ,  $\text{Ho}_2\text{O}_3$ ,  $\text{Er}_2\text{O}_3$ ,  $\text{Tm}_2\text{O}_3$  and  $\text{Yb}_2\text{O}_3$ , and  $\text{BaZrO}_3$ ,  $\text{MgO}$  and  $\text{MnO}$ , each having a purity of 99% or more, were prepared. These raw material powders and the oxides described above to be either one of the first or second side component were weighted so as to form compositions shown in TABLE 2002 and TABLE 2003. The amounts of addition of the first and second side components are indicated by parts by weight relative to 100 parts by weight of the essential component  $(\text{BaO})_m\text{TiO}_2 + \alpha\text{R}_2\text{O}_3 + \beta\text{BaZrO}_3 + \gamma\text{MgO} + \text{gMnO}$ .

Table 2002

\* indicates "out of the scope of the present invention"

Sam- ple No.	(BaO) <sub>m</sub> Kind of BaTiO <sub>3</sub>	TiO <sub>2</sub> + αR <sub>2</sub> O <sub>3</sub> + βBaZrO <sub>3</sub> + γMgO + gMnO												Amount of addition of the first compo- nent (parts by weight)	Amount of addi- tion of the se- cond side com- ponent (parts by weight)		
		α								Total of α	β	γ	g			γ + g	m
		Eu <sub>2</sub> O <sub>3</sub>	Gd <sub>2</sub> O <sub>3</sub>	Tb <sub>2</sub> O <sub>3</sub>	Dy <sub>2</sub> O <sub>3</sub>	Ho <sub>2</sub> O <sub>3</sub>	Er <sub>2</sub> O <sub>3</sub>	Tm <sub>2</sub> O <sub>3</sub>	Yb <sub>2</sub> O <sub>3</sub>								
*2001	2A	0	0.0008	0	0	0	0	0	0	0.0008	0.02	0.0015	0.0015	0.003	1.005	1	0
*2002	2A	0	0	0	0.04	0.03	0	0	0	0.07	0.03	0.04	0.09	0.13	1.01	1	0
*2003	2A	0	0.02	0.02	0	0	0	0	0	0.04	0	0.04	0.038	0.078	1.01	1.5	0
*2004	2A	0	0	0	0.03	0	0	0	0	0.03	0.08	0.03	0.02	0.05	1.01	1.5	0
*2005	2A	0.02	0	0	0	0	0	0	0	0.02	0.02	0.001	0.039	0.04	1.01	1	0
*2006	2A	0	0.02	0.03	0	0	0	0	0.01	0.06	0.02	0.125	0.005	0.13	1.01	1.5	0
*2007	2A	0	0	0	0	0.01	0	0.01	0	0.02	0.03	0.039	0.001	0.04	1.01	1	0
*2008	2A	0	0	0	0.04	0	0.02	0	0	0.06	0.02	0.005	0.125	0.13	1.01	1	0
*2009	2A	0	0.04	0.01	0.01	0	0	0	0	0.06	0.03	0.04	0.1	0.14	1.01	1	0
*2010	2A	0	0	0	0	0.02	0	0	0	0.02	0.03	0.02	0.03	0.05	0.99	1	0
*2011	2A	0.03	0	0	0	0	0	0	0	0.03	0.04	0.03	0.02	0.05	1.00	1	0
*2012	2A	0.01	0	0	0	0	0	0	0	0.01	0.04	0.015	0.005	0.02	1.038	1	0
*2013	2A	0	0.02	0	0	0	0	0	0	0.02	0.02	0.02	0.03	0.05	1.05	0	1
*2014	2A	0	0	0.02	0	0	0	0	0	0.02	0.02	0.02	0.01	0.03	1.01	0	0
*2015	2A	0	0	0.01	0.02	0	0	0	0	0.03	0.03	0.02	0.03	0.05	1.01	5	0
*2016	2A	0	0	0	0.02	0	0	0	0	0.02	0.02	0.03	0.01	0.04	1.01	0	0
*2017	2A	0	0.01	0	0	0	0	0	0	0.01	0.02	0.015	0.005	0.02	1.01	0	4
*2018	2D	0	0	0	0.03	0	0	0	0	0.03	0.03	0.03	0.02	0.05	1.01	2	0
2019	2A	0.02	0.01	0	0.02	0	0	0	0	0.05	0.02	0.05	0.07	0.12	1.015	1	0
2020	2B	0	0	0	0.02	0	0	0	0	0.02	0.03	0.02	0.01	0.03	1.02	1	0
2021	2C	0	0	0.02	0	0	0	0	0	0.06	0.03	0.12	0.002	0.122	1.03	1.5	0
2022	2A	0.05	0	0	0	0	0	0	0.01	0.06	0.03	0.06	0.07	0.13	1.02	1	0
2023	2A	0.01	0.01	0	0.04	0	0	0	0	0.06	0.03	0.002	0.12	0.122	1.01	0	1

Table 2003

Sam- ple No.	(BaO) <sub>m</sub> · Kind of BaTiO <sub>3</sub>	TiO <sub>2</sub> + αR <sub>2</sub> O <sub>3</sub> + βBaZrO <sub>3</sub> + γMgO + gMnO													The amount of addition of the first compo- nent (parts by weight)	The amount of addition of the second com- ponent (parts by weight)
		Total of α	β	γ	g	γ + g	m									
								Eu <sub>2</sub> O <sub>3</sub>	Gd <sub>2</sub> O <sub>3</sub>	Tb <sub>2</sub> O <sub>3</sub>	Dy <sub>2</sub> O <sub>3</sub>	Ho <sub>2</sub> O <sub>3</sub>	Er <sub>2</sub> O <sub>3</sub>	Tm <sub>2</sub> O <sub>3</sub>	Yb <sub>2</sub> O <sub>3</sub>	
2024	2A	0	0	0	0.001	0	0	0	0.001	0.02	0.0015	0.0015	0.003	1.01	1	0
2025	2A	0	0	0.01	0	0	0.01	0	0.02	0.02	0.02	0.03	0.05	1.01	1	0
2026	2A	0	0	0	0.02	0	0	0.01	0	0.03	0.02	0.03	0.05	1.015	0	1
2027	2A	0.02	0	0	0	0.02	0	0	0.04	0.02	0.03	0.05	0.08	1.01	0	1
2028	2A	0	0.05	0	0	0	0	0	0.05	0.02	0.05	0.06	0.11	1.01	1	0
2029	2A	0	0.02	0	0.04	0	0	0	0.06	0.02	0.06	0.062	0.122	1.01	1	0
2030	2A	0	0	0	0.01	0	0.01	0	0.02	0.005	0.02	0.03	0.05	1.01	1	0
2031	2A	0	0	0	0.01	0	0	0.01	0	0.02	0.06	0.02	0.04	1.01	1	0
2032	2A	0	0.02	0	0	0	0.01	0	0.03	0.03	0.02	0.04	0.06	1.035	2	0
2033	2A	0	0.01	0	0.01	0	0	0	0.02	0.03	0.02	0.03	0.05	1.015	0.2	0
2034	2A	0	0	0.01	0	0	0	0	0.01	0.02	0.01	0.012	0.022	1.01	3	0
2035	2A	0	0	0	0.01	0	0	0	0.01	0.03	0.015	0.005	0.02	1.01	0	0.2
2036	2A	0.01	0	0	0.01	0	0	0	0.02	0.03	0.02	0.03	0.05	1.01	0	3

Organic solvents such as polyvinyl butyral binder and ethanol were added to the weighed compounds, which were mixed in a ball mill in a wet state to prepare a ceramic slurry. This ceramic slurry was formed into a sheet by a doctor blade method to obtain a rectangular shaped green sheet with a thickness of 35 μm, followed by printing an electrocon-

ductive paste mainly composed of Ni on the ceramic green sheet to form an electroconductive paste layer for forming inner electrodes.

Then, a plurality of the ceramic green sheets on which the electroconductive layer is formed were laminated so that the sides where the electroconductive paste is projected out are alternately placed with each other, thus obtaining a monolithic body. This monolithic body was heated at 350 °C in a N<sub>2</sub> atmosphere and, after allowing the binder to decompose, the monolithic body was fired at the temperatures shown in TABLE 2004 and TABLE 2005 in a reducing atmosphere comprising H<sub>2</sub>-N<sub>2</sub>-H<sub>2</sub>O gases under an oxygen partial pressure of 10<sup>-9</sup> to 10<sup>-12</sup> MPa for two hours, thereby obtaining a ceramic sintered body.

The both side faces of the ceramic sintered body were coated with a silver paste containing B<sub>2</sub>O<sub>3</sub>-Li<sub>2</sub>O-SiO<sub>2</sub>-BaO glass frits and fired at a temperature of 600 °C in a N<sub>2</sub> atmosphere, thereby obtaining outer electrodes electrically connected to the inner electrodes.

The overall dimensions of the monolithic ceramic capacitor thus obtained were 5.0 mm in width, 5.7 mm in length and 2.4 mm in thickness while the thickness of the dielectric ceramic layer was 30 μm. Total number of the effective dielectric ceramic layers were 57, the area of the confronting electrode per one layer being  $8.2 \times 10^{-6} \text{ m}^2$ .

Electric characteristics of these monolithic ceramic capacitors were measured. The electrostatic capacitance (C) and dielectric loss (tan δ) were measured using an automatic bridge type measuring instrument at 1 kHz, 1 Vrms and 25 °C and the dielectric constant (ε) was calculated from the electrostatic capacitance. Next, the insulation resistance was measured using an insulation resistance tester at 25 °C and 150 °C by impressing direct current voltages of 315 V (or 10 kV/mm) and 945 V (or 30 kV/mm) for 2 minutes, obtaining a product of the electrostatic capacitance and insulation resistance, or a product CR.

The rate of change of the electrostatic capacitance against temperature changes was also measured. The rate of change at -25 °C and 85 °C by taking the electrostatic capacitance at 20 °C as a standard ( $\Delta C/C_{20}$ ), the rate of change at -55 °C and 125 °C by taking the electrostatic capacitance at 20 °C as a standard ( $\Delta C/C_{25}$ ) and the maximum value of the rate of change ( $|\Delta C| \text{ max}$ ) as an absolute value in the temperature range of -55 °C to 125 °C were measured as the electrostatic capacitances against temperature changes.

The DC bias characteristic was also evaluated. First, the electrostatic capacitance when an AC voltage of 1 kHz and 1 Vrms was impressed was measured. Then, the electrostatic capacitance when a DC voltage of 150 V and an AC voltage of 1 kHz and 1 Vrms were simultaneously impressed was measured, thereby the rate of reduction of the electrostatic capacitance ( $\Delta C/C$ ) due to loading the DC voltage was calculated.

In the high temperature load test, a direct current voltage of 750 V (or 25 kV/mm) was impressed at 150 °C on 36 pieces of each sample to measure the time dependent changes of the insulation resistance. The time when the insulation resistance of each sample was reduced below 10<sup>6</sup> Ω was defined to be a life span time and mean life span time was evaluated.

In the humidity resistance test, the number of the test pieces having an insulation resistance of 10<sup>6</sup> Ω or less among the 72 test pieces were counted after impressing a DC voltage of 315 V under an atmospheric pressure of 2 atm (relative humidity 100%) at 120 °C for 250 hours.

Insulation breakdown voltages under AC and DC voltages were measured by impressing AC and DC voltages at a voltage increase rate of 100 V/sec.

The results described above are listed in TABLE 2004 and TABLE 2005.

Table 2004

\* indicates "out of the scope of the present invention"

Sam- ple No.	Bak- ing temp. (°C)	Dielec- tric con- stant	Dielec- tric loss tan δ (%)	Ratio of temperature dependent capacitance change (%)			DC vias charac- teristic (%)	Product CR (Ω.F)				Insula- tion break- down voltage (kV/mm)		Humidi- ty resis- tance load test: Num- ber of injects	Mean life span (h)		
				Δ C/C <sub>20</sub>				Maxi- mum value	25°C				150°C				
				-25°C	85°C	-55°C			Δ C/C <sub>25</sub>	315V Im- pressed Voltage	945V Im- pressed Voltage	315V Im- pressed Voltage	945V Im- pressed Voltage			AC	DC
											Δ C/C	5kV/mm					
*2001	1300	2030	0.8	2.4	-8.6	4.3	-12.5	13.4	-56	3040	2890	110	110	13	14	0.72	850
*2002	1300	860	1	3.5	-14.9	6.5	-23.4	23.4	-14	5070	4820	240	230	13	14	0.72	170
*2003	1280	1350	0.9	3.6	-8.5	3.3	-13.4	13.4	-30	3020	2110	120	80	12	14	0.72	800
*2004	1300	1420	1	3.5	-13.2	4	-28.6	28.6	-36	5120	4860	210	200	12	14	0.72	120
*2005	1280	1550	0.9	3.6	-13.2	4	-17.2	17.2	-41	3040	2890	120	110	13	14	0.72	880
*2006	1360	1080	2.5	2.9	-9.1	4.5	-12.8	12.8	-16	5080	4830	220	210	12	14	49/72	180
*2007	Unmeasurable due to semiconductor formation																
*2008	1280	1060	0.8	4.2	-9.2	4.5	-18.3	18.3	-16	3060	2910	140	130	12	14	0.72	110
*2009	1280	1040	2.5	3.1	-9	4.1	-13.5	13.5	-15	5080	4830	230	220	12	14	15/72	100
*2010	Unmeasurable due to semiconductor formation																
*2011	1300	1460	0.9	3.4	-9.4	3.8	-13.4	13.4	-38	3180	3020	170	160	10	11	0.72	190
*2012	Unmeasurable due to insufficient sintering																
*2013	Unmeasurable due to insufficient sintering																
*2014	Unmeasurable due to insufficient sintering																
*2015	1300	1470	2.7	3.5	-9.3	4	-12.9	12.9	-39	3160	3000	180	170	10	12	0.72	150
*2016	Unmeasurable due to insufficient sintering																
*2017	1300	1920	2.6	3.2	-9.1	3.8	-12.7	12.7	-55	3250	3090	170	160	11	11	0.72	120
*2018	1300	1050	0.8	3	-8.7	4.2	-13.5	13.5	-15	5140	4880	250	240	13	15	0.72	850
2019	1280	1230	0.7	2.8	-8.9	3.8	-12.8	12.8	-22	5080	4830	260	250	12	15	0.72	890
2020	1280	1560	0.8	2.7	-9	3.8	-13.6	13.6	-42	5060	4810	270	260	12	14	0.72	920
2021	1280	1080	0.7	2.9	-9.2	3.7	-13.5	13.5	-16	5200	4940	280	270	12	15	0.72	890
2022	1300	1060	0.8	3	-9.4	3.9	-13.7	13.7	-16	5160	4900	260	250	12	14	0.72	850
2023	1300	1070	0.7	3.2	-9	4	-13.8	13.8	-16	5220	4960	250	240	13	15	0.72	900

due 2004

Table 2005

Sample No.	Baking temp. (°C)	Dielectric constant	Dielectric loss tan δ (%)	Ratio of temperature dependent capacitance change (%)					DC vias charac-teristic (%)	Product CR (Ω.F)				Insulation break-down voltage (kV/mm)		Humidity resis-tance load test: Num-ber of rejects	Mean life span (h)	
				Δ C/C <sub>25</sub>		Maximum value	Δ C/C <sub>25</sub>			150°C				AC	DC			
				-25°C	85°C		-55°C	125°C		315V Im-pressed Voltage	945V Im-pressed Voltage	315V Im-pressed Voltage	945V Im-pressed Voltage					
2024	1300	2010	0.9	3.4	-8.9	4	-13.6	14	-56	5260	5000	250	240	12	14	0/72	830	
2025	1300	1530	0.8	3.4	-8.7	4.1	-12.9	13	-42	5280	5020	280	270	12	14	0/72	840	
2026	1280	1470	0.8	3	-8.8	3.9	-13	13	-38	5100	4850	240	230	13	14	0/72	890	
2027	1280	1360	0.7	2.9	-8.9	3.8	-12.8	13	-32	5160	4900	260	250	12	14	0/72	820	
2028	1300	1220	0.7	3.5	-9.2	3.5	-13.2	13	-22	5290	5030	230	220	13	14	0/72	870	
2029	1300	1050	0.7	3.3	-9.3	3.8	-13.8	14	-15	5060	4810	210	200	12	14	0/72	900	
2030	1300	1550	0.7	3.2	-9.1	3.8	-13.5	14	-41	5120	4860	260	250	12	14	0/72	830	
2031	1280	1570	0.7	3	-9	4	-12.8	13	-41	5180	4920	250	240	12	15	0/72	810	
2032	1280	1430	0.7	3.5	-8.9	4.1	-13	13	-37	5280	5020	250	240	12	14	0/72	860	
2033	1300	1580	0.7	4	-8.8	3.9	-13.7	14	-41	5160	4900	230	220	12	14	0/72	810	
2034	1300	1600	0.7	4.1	-8.7	3.9	-13.5	14	-45	5090	4840	260	250	12	14	0/72	870	
2035	1300	1600	0.7	3.2	-9	3.5	-13.9	14	-45	5100	4850	250	240	13	14	0/72	850	
2036	1280	1540	0.7	3.5	-8.9	3.6	-12.9	13	-43	5160	4900	280	270	13	14	0/72	880	

It is evident from Table 2002 to TABLE 2005 that the monolithic ceramic capacitor according to the present invention has a capacitance decreasing ratio of as small as within -45% at an impressed voltage of 5 kV/mm and a dielectric loss of less than 1.0 %, wherein the rate of change against temperature changes satisfies both the B-level characteristic

standard stipulated in the JIS Standard in the temperature range of -25 °C to +85 °C and X7R-level characteristic standard stipulated in the EIA standard in the temperature range of -55 °C to +125 °C.

Moreover, the insulation resistances at 25 °C and 150 °C as expressed by the product CR show as high values as 5000  $\Omega \cdot F$  or more and 200  $\Omega \cdot F$  or more, respectively, when the ceramic capacitor is used under a high electric field strength of 10 kV/mm. The insulation breakdown voltage also shows high values of 12 kV/mm or more under the AC voltage and 14 kV/mm or more under the DC voltage. In addition, an acceleration test at 150 °C and DC 25 kV/mm gave a mean life span as long as 800 hours or more besides enabling a relatively low firing temperature of 1300 °C or less.

The reason why the composition was limited in the present invention will be described hereinafter.

In the composition of  $(\text{BaO})_m \text{TiO}_2 + \alpha \text{R}_2\text{O}_3 + \beta \text{BaZrO}_3 + \gamma \text{MgO} + g \text{MnO}$  (wherein  $\text{R}_2\text{O}_3$  represents at least one compound selected from  $\text{Eu}_2\text{O}_3$ ,  $\text{Gd}_2\text{O}_3$ ,  $\text{Tb}_2\text{O}_3$ ,  $\text{Dy}_2\text{O}_3$ ,  $\text{Ho}_2\text{O}_3$ ,  $\text{Er}_2\text{O}_3$ ,  $\text{Tm}_2\text{O}_3$  and  $\text{Yb}_2\text{O}_3$ ,  $\alpha$ ,  $\beta$ ,  $\gamma$ , and  $g$  representing mole ratio, respectively), the  $\text{R}_2\text{O}_3$  content  $\alpha$  of less than 0.001 as shown in the sample No. 2001 is not preferable because the insulation resistance is low and the product CR becomes small. On the other hand, the  $\text{R}_2\text{O}_3$  content  $\alpha$  of more than 0.06 as shown in the sample No. 2002 is also not preferable because the temperature characteristic does not satisfy the B-level characteristic / X7R characteristic, being poor in reliability. Accordingly, the preferable range of the  $\text{R}_2\text{O}_3$  content  $\alpha$  is  $0.001 \leq \alpha \leq 0.06$ .

The  $\text{BaZrO}_3$  content  $\beta$  of zero as in the sample No. 2003 is not preferable since the insulation resistance is low and the voltage dependency of the insulating resistance is larger than that of the composition system containing  $\text{BaZrO}_3$ . It is also not preferable that the  $\text{BaZrO}_3$  content  $\beta$  is more than 0.06 as in the sample No. 2004 because the temperature characteristic does not satisfy the B-level characteristic / X7R characteristic, along with shortening the mean life span. Accordingly, the preferable range of the  $\text{BaZrO}_3$  content  $\beta$  is  $0.005 \leq \beta \leq 0.06$ .

It is not preferable that, as seen in the sample No. 2005, the  $\text{MgO}$  content  $\gamma$  is 0.001 since the insulation resistance becomes low and the temperature characteristic does not satisfy the B-level characteristic / X7R characteristic. On the other hand, it is not preferable that the  $\text{MgO}$  content  $\gamma$  exceeds 0.12 as in the sample No. 2006, because the sintering temperature becomes high, the dielectric loss exceeds 2.0%, the number of rejections in the humidity resistance load test is extremely increased along with the mean life span being short. Accordingly, the preferable range of the  $\text{MgO}$  content  $\gamma$  is in the range of  $0.001 < \gamma \leq 0.12$ .

It is not preferable that, as seen in the sample No. 2007, the  $\text{MnO}$  content  $g$  is 0.001 since measurement becomes impossible due to formation of semiconductors. It is not preferable, on the other hand, that the  $\text{MnO}$  content  $g$  exceeds 0.12 as seen in the sample No. 2008 because the temperature characteristic X7R is not satisfied and the insulation resistance is lowered besides the mean life span becomes short. Accordingly, the preferable range of the  $\text{MnO}$  content  $g$  is in the range of  $0.001 < g \leq 0.12$ .

It is not preferable that, as in the sample No. 2009, the combined amount of  $\gamma + g$  of the  $\text{MgO}$  content and  $\text{MnO}$  content exceeds 0.13 because the dielectric loss is increased to 2.0% and the mean life span is shortened besides the rejection number in the humidity resistance load test increases. Accordingly, the combined amount of  $\gamma + g$  of the  $\text{MgO}$  content and  $\text{MnO}$  content is preferably in the range of  $\gamma + g \leq 0.13$ .

It is not preferable that the  $\text{BaO}/\text{TiO}_2$  ratio  $m$  is less than 1.000 as in the sample No. 2010 because measurements are impossible due to formation of semiconductors. It is also not preferable that, as seen in the sample No. 2011, the  $\text{BaO}/\text{TiO}_2$  ratio  $m$  is 1.000 since the insulation resistance as well as the AC and DC breakdown voltage becomes low along with shortening the mean life span. It is not preferable, on the other hand, that the  $\text{BaO}/\text{TiO}_2$  ratio  $m$  is over 1.035 as in the samples No. 2012 and 2013 since measurements becomes impossible due to insufficient sintering. Accordingly, the  $\text{BaO}/\text{TiO}_2$  ratio  $m$  in the range of  $1.000 < m \leq 1.035$  is preferable.

It is not preferable that the amount of addition of the first or second side component is zero as in the samples No. 2014 and 2016 because measurements are impossible due to insufficient sintering. It is not preferable that the amount of addition of the first or second side component exceeds 3.0 parts by weight as seen in the samples No. 2015 and 2017, on the other hand, because the dielectric loss exceeds 1.0% and the insulation resistance and insulation breakdown voltage are lowered along with shortening the mean life span. Accordingly, the preferable content of either the first or the second components is 0.2 to 3.0 parts by weight.

The contents of the alkali earth metal oxides contained in barium titanate as impurities are suppressed below 0.02% by weight because, when the contents of the alkali earth metal oxides exceeds 0.02% by weight as in the sample No. 2018, the dielectric constant is decreased.

#### (Example 14)

A material with a composition of  $\text{BaO}_{1.010} \cdot \text{TiO}_2 + 0.03\text{Gd}_2\text{O}_3 + 0.025\text{BaZrO}_3 + 0.05\text{MgO} + 0.01 \text{MnO}$  (mole ratio) was prepared using barium titanate in TABLE 1A as a dielectric powder. A monolithic ceramic capacitor was produced by the same method as in Example 1, except that an oxide represented by  $\text{Li}_2\text{O}-(\text{Si}, \text{Ti})\text{O}_2-\text{MO}$  shown in Table 2006, having a mean particle size of 1  $\mu\text{m}$  or less produced by heating the material described above at 1200 to 1500 °C, was added as the first side component. The overall dimensions of the monolithic ceramic capacitor produced is the same as

in Example 1.

Table 2006

Sample No.	Amount of addition (parts by weight)	The first side component				
		Composition (mol%, except w)				
		Li <sub>2</sub> O	(Si <sub>w</sub> Ti <sub>1-w</sub> )O <sub>2</sub>	w	Al <sub>2</sub> O <sub>3</sub>	ZrO <sub>2</sub>
2101	1	20	80	0.3	0	0
2102	1	10	80	0.6	5	5
2103	0.8	10	70	0.5	20	0
2104	0.8	35	45	1	10	10
2105	1.5	45	45	0.5	10	0
2106	1.5	45	55	0.3	0	0
2107	1	20	70	0.6	5	5
2108	1	20	70	0.4	10	0
2109	1.2	30	60	0.7	5	5
2110	1.2	30	60	0.8	10	0
2111	2	40	50	0.6	5	5
2112	2	40	50	0.9	0	10
2113	1.5	10	85	0.4	5	0
2114	2	5	75	0.6	10	10
2115	1.2	20	55	0.5	25	0
2116	1	45	40	0.8	0	15
2117	0.8	50	45	0.7	5	0
2118	1.2	25	75	0.9	0	0
2119	1.5	25	75	1	0	0
2120	1	35	65	0.9	0	0
2121	1.5	35	65	1	0	0
2122	1.2	20	70	0.2	0	10

The electric characteristics were then measured by the same method as in Example 1. The results are shown in TABLE 2007.



Table 2007

Sample No.	Baking temp. (°C)	Dielectric constant	Dielectric loss tan δ (%)	Ratio of temperature dependent capacitance change (%)					DC vias characteristic (%)	Product CR (Ω.F)				Insulation breakdown voltage (kV/mm)		Humidity resistance load test: Number of rejections	Mean life span (h)	
				ΔC/C <sub>20</sub>	85°C	-55°C	ΔC/C <sub>25</sub>	125°C		Maximum value	ΔC/C	5kV/mm	25°C		150°C			
2101	1280	1430	0.8	3.6	-9	4.2	-13.4	13	-37	5240	4980	240	230	230	12	14	0/72	850
2102	1280	1460	0.8	3.8	-8.7	3.8	-13.5	14	-37	5130	4870	220	210	210	12	14	0/72	890
2103	1280	1490	0.8	4.1	-8.9	3.9	-13.6	14	-40	5170	4910	220	210	210	12	14	0/72	870
2104	1300	1420	0.8	4	-9	4	-14	14	-35	5090	4840	230	220	220	12	14	0/72	830
2105	1300	1430	0.9	3.2	-9.2	4.2	-13.8	14	-36	5080	4830	220	210	210	13	14	0/72	840
2106	1280	1400	0.8	3.6	-9.4	3.7	-13.7	14	-35	5100	4850	210	200	200	12	14	0/72	850
2107	1280	1460	0.8	3.5	-8.8	3.5	-13.9	14	-38	5160	4900	210	200	200	13	14	0/72	890
2108	1280	1460	0.8	4	-9.5	3.9	-12.8	13	-38	5240	4980	210	200	200	12	15	0/72	840
2109	1280	1420	0.9	3.8	-9.6	4.2	-12.5	13	-36	5230	4970	210	200	200	12	14	0/72	830
2110	1300	1470	0.8	3.5	-9.7	3.5	-13	13	-38	5200	4940	210	200	200	12	14	0/72	840
2111	1300	1430	0.8	3.9	-9.5	4.2	-13.8	14	-35	5180	4920	220	210	210	13	14	0/72	890
2112	1280	1420	0.8	4	-9.6	4.3	-13.6	14	-35	5270	5010	220	210	210	12	14	0/72	900
2113	1350	Unmeasurable due to insufficient sintering																
2114	1350	Unmeasurable due to insufficient sintering																
2115	1350	1230	1.8	3.5	-9.1	4	-13.8	14	-22	3800	3610	150	140	140	11	12	26/72	150
2116	1350	Unmeasurable due to insufficient sintering																
2117	1350	Unmeasurable due to insufficient sintering																
2118	1300	1480	0.9	3.8	9.3	4.6	-13.5	14	-37	5160	4900	220	210	210	12	14	0/72	860
2119	1350	1200	1.6	3.2	-8.8	3.8	-12.9	13	-20	3860	3670	120	110	110	11	12	20/72	160
2120	1300	1450	0.9	3	-8.9	3.8	-13.7	14	-38	5230	4970	230	220	220	12	14	0/72	840
2121	1350	1210	1.4	3.8	-8.9	3.5	-13.6	14	-21	3810	3620	130	120	120	11	12	34/72	130
2122	1350	1190	1.5	3.6	-9	4	-13.5	14	-20	3850	3660	100	90	90	11	12	29/72	180

As is evident from TABLE 2006 and TABLE 2007, preferable results are obtained in the samples No. 2101 to 2112, 2118 and 2120 in which the oxides with compositions within or on the boundary lines of the area surrounded by the

straight lines connecting each spot indicated by A ( $X = 20, y = 80, z = 0$ ), B ( $X = 10, y = 80, z = 10$ ), C ( $X = 10, y = 70, z = 20$ ), D ( $X = 35, y = 45, z = 20$ ), E ( $x = 45, y = 45, z = 10$ ) and F ( $x = 45, y = 55, z = 0$ ) (wherein  $x, y$  and  $z$  represent mole % and  $w$  represents mole ratio,  $w$  being in the range of  $0.3 \leq w < 1.0$  when it falls on the line A - F) of the three component phase diagram of the oxides represented by  $\text{Li}_2\text{O}-(\text{Si}_w\text{Ti}_{1-w})\text{O}_2\text{-MO}$  shown in FIG. 4 are added, wherein the samples have a capacitance decreasing ratio of as small as within -45% at an impressed voltage of 5 kV/mm and a dielectric loss of 1.0% or less, along with the rate of change of the electrostatic capacitance against temperature changes satisfying the B-level characteristic standard stipulated in the JIS Standard in the temperature range of -25 °C to +85 °C and X7R-level characteristic standard stipulated in the EIA standard in the temperature range of -55 °C to +125 °C.

Moreover, the insulation resistances at 25 °C and 150 °C as expressed by the product CR show as high values as 5000  $\Omega \cdot \text{F}$  or more and 200  $\Omega \cdot \text{F}$  or more, respectively, when the ceramic capacitor is used under a high electric field strength of 10 kV/mm. The insulation breakdown voltage also shows high values of 12 kV/mm or more under the AC voltage and 14 kV/mm or more under the DC voltage. In addition, an acceleration test at 150 °C and DC 25 kV/mm gave a mean life span as long as 800 hours or more besides enabling a relatively low firing temperature of 1300 °C or less.

When the oxide represented by  $\text{Li}_2\text{O}-(\text{Si}_w\text{Ti}_{1-w})\text{O}_2\text{-MO}$  has a composition outside of the composition described above as in the samples No. 2113 to 2117 and 2119, on the other hand, sintering becomes insufficient or many rejections appear in the humidity resistance load test even after sintering. When the composition falls on the line A - F and  $w = 1.0$ , the sintering temperature becomes high, giving a lot of rejections in the humidity resistance load test as shown in the samples No. 2119 and 2121. When the value of  $w$  is less than 3.0, the sintering temperature becomes so high that many rejections appear in the humidity resistance test as shown in Sample No. 2122.

#### (Example 15)

A material with a composition of  $\text{BaO}_{1.010} \cdot \text{TiO}_2 + 0.03\text{Dy}_2\text{O}_3 + 0.02\text{BaZrO}_3 + 0.05 \text{MgO} + 0.01 \text{MnO}$  (mole ratio) was prepared using barium titanate in TABLE 1A as a dielectric powder. A monolithic ceramic capacitor was produced by the same method as in Example 1, except that an oxide represented by  $\text{SiO}_2\text{-TiO}_2\text{-XO}$  shown in Table 2008, having a mean particle size of 1  $\mu\text{m}$  or less produced by heating the material described above at 1200 to 1500 °C, was added as the second side component. The amounts of addition of  $\text{Al}_2\text{O}_3$  and  $\text{ZrO}_3$  are indicated by parts by weight relative to 100 parts by weight of the second side component ( $x\text{SiO}_2\text{-yTiO}_2\text{-zXO}$ ). The overall dimensions of the monolithic ceramic capacitor produced is the same as in Example 1.

Table 2008

Sample No.	The second side component														Added component (parts by weight)	
	Amount of addition (parts by weight)	Essential component (mol %)														
		SiO <sub>2</sub>	TiO <sub>2</sub>	XO							Total					
				BaO	CaO	SrO	MgO	ZnO	MnO							
2201	1	85	1	1	0	0	0	4	9	14	0	0	0			
2202	1	35	51	0	10	0	0	0	4	14	0	0	0			
2203	1	30	20	0	30	0	15	4	1	50	0	0	0			
2204	1	39	1	20	20	2	0	13	5	60	0	0	0			
2205	1	70	10	5	5	0	0	10	0	20	0	0	0			
2206	1	45	10	0	0	0	0	15	30	45	0	0	0			
2207	1	50	20	10	10	3	7	0	0	30	0	0	0			
2208	1	50	30	0	16	0	0	0	4	20	0	0	0			
2209	1	35	30	25	10	0	0	0	0	35	0	0	0			
2210	1	40	40	10	0	0	0	5	5	20	0	0	0			
2211	1	45	22	3	30	0	0	0	0	33	15	0	0			
2212	1	45	22	3	30	0	0	0	0	33	10	5	0			
2213	1	65	25	5	5	0	0	0	0	10	0	0	0			
2214	1	25	40	15	0	10	0	5	5	35	0	0	0			
2215	1	30	10	30	25	0	0	5	0	60	0	0	0			
2216	1	50	0	35	15	0	0	0	0	50	0	0	0			
2217	1	45	22	30	0	0	3	0	0	33	25	0	0			
2218	1	45	22	30	0	3	0	0	0	33	0	15	0			
2219	1	30	60	10	0	0	0	0	0	10	0	0	0			

The electric characteristics were then measured by the same method as in Example 1. The results are shown in TABLE 2009.

Table 2009

Sample No.	Baking temp. (°C)	Dielectric constant	Dielectric loss tan δ (%)	Ratio of temperature dependent capacitance change (%)					DC vias characteristic (%)	Product CR (Ω·F)				Insulation breakdown voltage (kV/mm)		Humidity resistance load test: Number of rejects	Mean life span (h)	
				Δ C/C <sub>20</sub>	85°C	-55°C	Δ C/C <sub>25</sub>	125°C		Maximum value	315V Impressed Voltage	945V Impressed Voltage	315V Impressed Voltage	945V Impressed Voltage	150°C			AC
2201	1300	1040	0.7	-25°C	3.4	-9.5	3.6	-13.9	14	-16	5070	4820	210	200	13	15	0/72	880
2202	1280	1060	0.7		3.5	-8.9	4	-14.2	14	-17	5090	4840	210	200	12	14	0/72	850
2203	1280	1100	0.7		3.8	-9.1	3.9	-13.8	14	-18	5100	4850	220	210	12	15	0/72	800
2204	1300	1070	0.7		4	-9	4.2	-13.5	14	-17	5120	4860	210	200	12	14	0/72	900
2205	1300	1020	0.7		4.2	-9.2	4.3	-13.6	14	-15	5130	4870	210	200	13	14	0/72	920
2206	1280	1080	0.7		3.9	-8.8	4.5	-13.7	14	-17	5080	4830	230	220	13	15	0/72	840
2207	1280	1100	0.7		3.4	-8.7	4.5	-14	14	-18	5080	4830	210	200	12	14	0/72	860
2208	1300	1060	0.7		3.6	-8.9	4.2	-13.5	14	-17	5120	4860	220	210	12	14	0/72	920
2209	1300	1090	0.7		4.1	-8.8	4	-13.6	14	-18	5160	4900	220	210	12	14	0/72	900
2210	1300	1050	0.7		3.8	-9	4.3	-13.7	14	-17	5150	4890	210	200	12	14	0/72	880
2211	1280	1070	0.7		3.9	-9.2	4.2	-13.3	13	-17	5430	5160	310	300	12	15	0/72	870
2212	1300	1080	0.7		4	-9.4	4	-13.5	14	-18	5450	5180	320	300	12	14	0/72	900
2213	1350	860	1.4		3.5	-8.7	3.9	-13.6	14	-14	3790	3600	150	140	11	13	45/72	160
2214	1350	Unmeasurable due to insufficient sintering																
2215	1350	Unmeasurable due to insufficient sintering																
2216	1350	830	1.3	3.6	-8.8	3.9	-13.7	14	-14	-14	3860	3670	130	120	11	12	68/72	180
2217	1350	Unmeasurable due to insufficient sintering																
2218	1350	Unmeasurable due to insufficient sintering																
2219	1350	Unmeasurable due to insufficient sintering																

As is evident from TABLE 2008 and TABLE 2009, preferable results are obtained in the samples No. 2201 to 2212, in which oxides with compositions within or on the boundary lines of the area surrounded by the straight lines connecting each spot indicated by A ( $x = 85$ ,  $y = 1$ ,  $z = 14$ ), B ( $X = 35$ ,  $y = 51$ ,  $z = 14$ ), C ( $X = 30$ ,  $y = 20$ ,  $z = 50$ ) and D ( $X = 39$ ,

y = 1, z = 60), (wherein x, y and z represent mole %), of the three component phase diagram of the oxides represented by  $\text{SiO}_2\text{-TiO}_2\text{-XO}$  shown in FIG. 5 are added, wherein the samples have a capacitance decreasing ratio of as small as within -45% at an impressed voltage of 5 kV/mm and a dielectric loss of 1.0% or less, along with the rate of change of the electrostatic capacitance against temperature changes satisfying the B-level characteristic standard stipulated in the JIS Standard in the temperature range of -25 °C to +85 °C and X7R-level characteristic standard stipulated in the EIA standard in the temperature range of -55 °C to +125 °C.

Moreover, the insulation resistances at 25 °C and 150 °C as expressed by the product CR show as high values as 5000  $\Omega \cdot \text{F}$  or more and 200  $\Omega \cdot \text{F}$  or more, respectively, when the ceramic capacitor is used under a high electric field strength of 10 kV/mm. The insulation breakdown voltage also shows high values of 12 kV/mm or more under the AC voltage and 14 kV/mm or more under the DC voltage. In addition, an acceleration test at 150 °C and DC 25 kV/mm gave a mean life span as long as 800 hours or more and no rejections were found in the humidity resistance load test besides enabling a relatively low firing temperature of 1300 °C or less.

When the oxide represented by  $\text{SiO}_2\text{-TiO}_2\text{-XO}$  has a composition outside of the composition described above as in the samples No. 2213 to 2219, on the contrary, sintering becomes insufficient or many rejection appear in the humidity resistance load test even after sintering.

While a monolithic capacitor having an insulation resistance of 5400  $\Omega \cdot \text{F}$  or more and 300  $\Omega \cdot \text{F}$  or more at 25 °C and 150 °C, respectively, under a strong electric field of 10 kV/mm can be obtained by allowing  $\text{Al}_2\text{O}_3$  and/or  $\text{ZrO}_2$  to contain in the  $\text{SiO}_2\text{-TiO}_2\text{-XO}$  oxides as in the sample No. 2211 and 2212, sintering property is extremely decreased when  $\text{Al}_2\text{O}_3$  and  $\text{ZrO}_2$  are added in an amounts of 15 parts by weight or more and 5 parts by weight or more, respectively, as in the samples No. 2217 and 2218.

(Example 16)

Four kinds of barium titanate ( $\text{BaTiO}_3$ ), an oxide powder as a first side component and an oxide powder as a second side component were obtained by the same method as in Example 1.

Then,  $\text{BaCO}_3$  for adjusting the mole ratio Ba/Ti in barium titanate,  $\text{Eu}_2\text{O}_3$ ,  $\text{Gd}_2\text{O}_3$ ,  $\text{Tb}_2\text{O}_3$ ,  $\text{Dy}_2\text{O}_3$ ,  $\text{Ho}_2\text{O}_3$ ,  $\text{Er}_2\text{O}_3$ ,  $\text{Tm}_2\text{O}_3$  and  $\text{Yb}_2\text{O}_3$ , and  $\text{MnO}$ , each having a purity of 99% or more, were prepared. These raw material powders and the oxides described above to be either the first or the second component were weighed so as to be the composition in TABLE 2010 and TABLE 2011. The amounts of addition of the first and second side components are defined by the amount of addition relative to 100 parts by weight of  $(\text{BaO})_m\text{TiO}_2 + \alpha\text{R}_2\text{O}_3 + \beta\text{BaZrO}_3 + \gamma\text{MnO}$ . A monolithic ceramic capacitor was produced by the same method as in Example 1 using these weighed materials. The overall dimensions of the monolithic ceramic capacitor are the same as in Example 1.

Table 2010

\*indicates "out of the scope of the present invention"

Sample No.	(BaO) <sub>m</sub> · TiO <sub>2</sub> + αR <sub>2</sub> O <sub>3</sub> + βBaZrO <sub>3</sub> + γMnO	α								Total of α	β	γ	m	Amount of addition of the first component (parts by weight)	Amount of addition of the second component (parts by weight)	
		Kind of BaTiO <sub>3</sub>														
			Eu <sub>2</sub> O <sub>3</sub>	Gd <sub>2</sub> O <sub>3</sub>	Tb <sub>2</sub> O <sub>3</sub>	Dy <sub>2</sub> O <sub>3</sub>	Ho <sub>2</sub> O <sub>3</sub>	Er <sub>2</sub> O <sub>3</sub>	Tm <sub>2</sub> O <sub>3</sub>							Yb <sub>2</sub> O <sub>3</sub>
*2301	A	0	0.0007	0	0	0	0	0	0	0.0007	0.03	0.0014	1.005	1	0	
*2302	A	0	0	0.02	0	0.055	0	0	0	0.075	0.02	0.128	1.01	1	0	
*2303	A	0.01	0	0	0.025	0	0	0	0	0.035	0	0.07	1.015	2	0	
*2304	A	0	0.01	0	0	0.01	0	0.01	0	0.03	0.07	0.06	1.01	2.5	0	
*2305	A	0.04	0	0	0	0	0	0	0	0.04	0.025	0.001	1.01	1	0	
*2306	A	0	0	0	0.02	0	0.03	0	0	0.05	0.03	0.145	1.01	1	0	
*2307	A	0	0	0.02	0	0	0	0	0.01	0.03	0.03	0.06	0.99	0	1	
*2308	A	0	0.01	0	0.01	0	0	0	0	0.02	0.03	0.04	1	2	0	
*2309	A	0	0	0	0	0	0	0.01	0	0.01	0.03	0.02	1.037	1	0	
*2310	A	0.01	0.01	0	0	0	0	0	0	0.02	0.03	0.04	1.045	0	2	
*2311	A	0	0	0	0	0.01	0	0	0	0.01	0.02	0.02	1.01	0	0	
*2312	A	0	0.02	0	0	0	0.02	0	0	0.04	0.03	0.08	1.01	4	0	
*2313	A	0	0	0.01	0.01	0	0	0	0	0.02	0.04	0.04	1.015	0	0	
*2314	A	0.01	0	0	0	0	0	0	0.01	0.02	0.02	0.04	1.01	0	5	
*2315	D	0	0	0	0	0.02	0	0	0	0.02	0.04	0.04	1.01	2	0	
2316	A	0	0.02	0.02	0	0	0	0	0	0.04	0.02	0.08	1.01	0	1	
2317	B	0	0	0.01	0	0	0.01	0	0	0.02	0.03	0.04	1.02	1	0	

Table 2011

Sam- ple No.	(BaO) <sub>m</sub> · TiO <sub>2</sub> + αR <sub>2</sub> O <sub>3</sub> + βBaZrO <sub>3</sub> + γMnO												Amount of addition of the first side compo- nent (parts by weight)	Amount of addition of the second side compo- nent (parts by weight)		
	Kind of BaTiO <sub>3</sub>	α								Total of α	β	γ			m	
		Eu <sub>2</sub> O <sub>3</sub>	Gd <sub>2</sub> O <sub>3</sub>	Tb <sub>2</sub> O <sub>3</sub>	Dy <sub>2</sub> O <sub>3</sub>	Ho <sub>2</sub> O <sub>3</sub>	Er <sub>2</sub> O <sub>3</sub>	Tm <sub>2</sub> O <sub>3</sub>	Yb <sub>2</sub> O <sub>3</sub>							
2318	C	0.03	0	0	0	0	0	0	0	0	0.03	0.02	0.13	1.03	2	0
2319	A	0	0	0	0.01	0	0	0.02	0		0.03	0.02	0.06	1.015	1	0
2320	A	0	0.04	0	0	0	0	0	0	0	0.04	0.03	0.08	1.02	0	2
2321	A	0	0.001	0	0	0	0	0	0	0	0.001	0.02	0.002	1.01	1	0
2322	A	0	0	0.01	0.01	0	0	0	0.01	0	0.03	0.03	0.06	1.01	1	0
2323	A	0	0	0	0	0	0.02	0	0	0	0.02	0.02	0.04	1.025	0	1
2324	A	0	0.01	0	0	0.03	0	0	0	0	0.04	0.02	0.08	1.01	0	1
2325	A	0	0	0	0.02	0	0	0.02	0	0	0.04	0.03	0.08	1.01	1	0
2326	A	0	0.03	0	0	0	0.03	0	0	0	0.06	0.03	0.12	1.02	2	0
2327	A	0	0	0.01	0	0	0	0.02	0	0	0.03	0.005	0.06	1.01	1	0
2328	A	0	0	0	0.01	0.01	0	0	0	0	0.02	0.06	0.04	1.01	1	0
2329	A	0	0	0.01	0	0	0.01	0	0	0	0.02	0.02	0.04	1.035	2	0
2330	A	0	0	0	0	0	0	0.03	0	0	0.03	0.03	0.06	1.01	0.2	0
2331	A	0.01	0	0.01	0	0	0	0	0	0	0.02	0.02	0.04	1.01	3	0
2332	A	0	0	0	0.01	0	0	0	0.01	0	0.02	0.04	0.04	1.01	0	0.2
2333	A	0	0	0	0	0	0.02	0	0	0	0.02	0.03	0.04	1.01	0	3

The electric characteristics were measured by the same method as in Example 1. The results are shown in TABLE 2012 and TABLE 2013.

Table 2012

\* indicates "out of the scope of the present invention"

Sample No.	Baking temp. (°C)	Dielectric constant	Dielectric loss tan δ (%)	Ratio of temperature dependent capacitance change (%)				DC bias charac-teristic (%) ΔC/C 5kV/mm	Product CR (Ω · F)				Insulation breakdown voltage (kV/mm)		Humidity resis-tance load test: Number of rejects	Mean life span (h)			
				ΔC/C <sub>20</sub>		Maxi-mum value	315V Im-pressed Voltage		945V Im-pressed Voltage	315V Im-pressed Voltage	945V Im-pressed Voltage	AC	DC						
				-25°C	85°C									-55°C			125°C		
																		25°C	150°C
*2301	1300	1560	0.8	4	-13	5	-18	18	5200	4940	230	220	12	14	0/72	880			
*2302	1300	810	0.7	2.4	-7	4	-7	7	5430	5160	200	190	12	14	12/72	920			
*2303	1300	1330	0.8	2.2	-7.8	5	-8	8	3000	2100	90	60	12	14	0/72	860			
*2304	1300	1330	0.7	2.4	-14.2	4.8	-27	27	5230	4970	220	210	12	14	0/72	130			
*2305	Unmeasurable due to semiconductor formation																		
*2306	1280	1120	0.8	3.4	-8.3	4.6	-19	19	3080	2930	130	120	12	14	0/72	120			
*2307	Unmeasurable due to semiconductor formation																		
*2308	1300	1440	0.7	3.2	-8.2	4.1	-9	9	3200	3040	120	110	10	12	0/72	110			
*2309	Unmeasurable due to insufficient sintering																		
*2310	Unmeasurable due to insufficient sintering																		
*2311	Unmeasurable due to insufficient sintering																		
*2312	1300	1280	2.2	3.3	-8.7	4.3	-9.2	9.2	3310	3150	130	120	11	11	0/72	140			
*2313	Unmeasurable due to insufficient sintering																		
*2314	1300	1420	2.4	2.7	-7.6	3.6	-8	8	3200	3040	150	140	10	11	0/72	170			
*2315	1300	1120	0.7	2.4	-7.2	5	-9.1	9.1	5240	4980	210	200	12	14	0/72	900			
2316	1280	1220	0.7	2.4	-8.4	4.5	-11	11	5130	4870	250	240	12	14	0/72	850			
2317	1280	1450	0.7	2.6	-8.6	4.6	-12.1	12.1	5160	4900	230	220	13	15	0/72	890			



Table 2013

Sam- ple No.	Baking temp. (°C)	Dielec- tric con- stant	Dielec- tric loss tan δ (%)	Ratio of temperature dependent capacitance change (%)				DC bias charac- teristic (%)	Product CR (Ω · F)				Insulation breakdown voltage (kV/mm)		Humidity resistance load test: Number of rejection	Mean life span (h)						
				ΔC/C <sub>20</sub>		ΔC/C <sub>25</sub>			Maxi- mum value	315V Im- pressed Voltage	945V Im- pressed Voltage	315V Im- pressed Voltage	945V Im- pressed Voltage	AC			DC					
						-25°C	85°C											-55°C	125°C			
													25°C				150°C					
2318	1280	1360	0.8	3	-9	4.7	-10.4	10.4	-28	5200	4940	220	210	12	14	0/72	930					
2319	1300	1370	0.7	3.4	-9.1	4.5	-9.7	9.7	-30	5080	4830	240	230	12	14	0/72	870					
2320	1300	1240	0.7	2.8	-8.7	5	-9.5	9.5	-22	5140	4880	230	220	12	14	0/72	830					
2321	1300	1510	0.7	2.6	-8.5	4.8	-13.4	13.4	-39	5300	5040	230	220	12	14	0/72	950					
2322	1300	1360	0.7	2	-9	4.6	-10.3	10.3	-29	5220	4960	210	200	12	14	0/72	880					
2323	1300	1460	0.7	3	-9.2	4.9	-11.2	11.2	-38	5290	5030	250	240	12	14	0/72	900					
2324	1280	1250	0.8	3.2	-8.8	4.6	-11	11	-23	5340	5070	230	220	13	15	0/72	860					
2325	1300	1240	0.7	3.3	-8.7	4.3	-12	12	-23	5330	5060	220	210	12	14	0/72	830					
2326	1300	960	0.7	2.9	-9.2	5.1	-12.5	12.5	-12	5080	4830	200	190	13	15	0/72	810					
2327	1300	1340	0.7	2.8	-9.3	5.3	-12.1	12.1	-30	5210	4950	210	200	12	14	0/72	870					
2328	1280	1440	0.7	2.1	-8.5	4	-11.5	11.5	-38	5240	4980	220	210	12	14	0/72	910					
2329	1300	1470	0.8	2.6	-8.2	4.6	-13.6	13.6	-39	5260	5000	230	220	12	14	0/72	950					
2330	1300	1360	0.7	3	-8.8	4.2	-11.8	11.8	-28	5220	4960	220	210	12	14	0/72	880					
2331	1280	1420	0.8	2.7	-9	4.8	-11.9	11.9	-36	5240	4980	210	200	12	14	0/72	930					
2332	1300	1430	0.7	2.8	-8.6	4.7	-11	11	-36	5280	5020	230	220	12	14	0/72	910					
2333	1300	1460	0.7	3	-9.1	4.3	-10.7	10.7	-39	5300	5040	220	210	12	14	0/72	860					

It is evident from Table 2010 to TABLE 2013 that the monolithic ceramic capacitor according to the present invention has a capacitance decreasing ratio of as small as within -40% or less at an impressed voltage of 5 kV/mm and a

dielectric loss of less than 1.0 %, wherein the rate of change against temperature changes satisfies both the B-level characteristic standard stipulated in the JIS Standard in the temperature range of -25 °C to +85 °C and X7R-level characteristic standard stipulated in the EIA standard in the temperature range of -55 °C to +125 °C.

Moreover, the insulation resistances at 25 °C and 150 °C as expressed by the product CR show as high values as 5000  $\Omega \cdot F$  or more and 200  $\Omega \cdot F$  or more, respectively, when the ceramic capacitor is used under a high electric field strength of 10 kV/mm. The insulation breakdown voltage also shows high values of 12 kV/mm or more under the AC voltage and 14 kV/mm or more under the DC voltage. In addition, an acceleration test at 150 °C and DC 25 kV/mm gave a mean life span as long as 800 hours or more besides enabling a relatively low firing temperature of 1300 °C or less.

The reason why the composition was limited in the present invention will be described hereinafter.

In the composition of  $(\text{BaO})_m\text{TiO}_2 + \alpha\text{R}_2\text{O}_3 + \beta\text{BaZrO}_3 + \gamma\text{MgO}$  (wherein  $\text{R}_2\text{O}_3$  represents at least one compound selected from  $\text{Eu}_2\text{O}_3$ ,  $\text{Gd}_2\text{O}_3$ ,  $\text{Tb}_2\text{O}_3$ ,  $\text{Dy}_2\text{O}_3$ ,  $\text{Ho}_2\text{O}_3$ ,  $\text{Er}_2\text{O}_3$ ,  $\text{Tm}_2\text{O}_3$  and  $\text{Yb}_2\text{O}_3$ ,  $\alpha$ ,  $\beta$  and  $\gamma$  representing mole ratio, respectively), the  $\text{Mn}_2\text{O}_3$  content  $\alpha$  of less than 0.001 as shown in the sample No. 2301 is not preferable because the temperature characteristics does not satisfy the B-level characteristics / X7R characteristics. On the other hand, the  $\text{Mn}_2\text{O}_3$  content  $\alpha$  of more than 0.06 as shown in the sample No. 2302 is also not preferable because the specific dielectric constant becomes as small as less than 900. Accordingly, the preferable range of the  $\text{Mn}_2\text{O}_3$  content  $\alpha$  is  $0.001 \leq \alpha \leq 0.06$ .

The  $\text{BaZrO}_3$  content  $\beta$  of zero as in the sample No. 2303 is not preferable since the insulation resistance is low and the voltage dependency of the insulating resistance is larger than that of the composition system containing  $\text{BaZrO}_3$ . It is also not preferable that the  $\text{BaZrO}_3$  content  $\beta$  is more than 0.06 as in the sample No. 2304 because the temperature characteristic does not satisfy the B-level characteristic / X7R characteristic, along with shortening the mean life span. Accordingly, the preferable range of the  $\text{BaZrO}_3$  content  $\beta$  is  $0.005 \leq \beta \leq 0.06$ .

It is not preferable that, as seen in the sample No. 2305, the  $\text{MgO}$  content  $\gamma$  is 0.001 since measuring is impossible due to formation of semiconductors. On the other hand, it is not preferable that the  $\text{MgO}$  content  $\gamma$  exceeds 0.13 as in the sample No. 2306, because the temperature characteristic X7R is not satisfied and the insulation capacitance is low along with the mean life span being short. Accordingly, the  $\text{Mn}$  content  $\gamma$  is preferably in the range of  $0.001 \leq \gamma < 0.13$ .

It is not preferable that the  $\text{BaO}/\text{TiO}_2$  ratio  $m$  is less than 1.000 as in the sample No. 2307 because measurements are impossible due to formation of semiconductors. It is also not preferable that, as seen in the sample No. 2308, the  $\text{BaO}/\text{TiO}_2$  ratio  $m$  is 1.000 since the insulation resistance as well as the AC and DC breakdown voltage becomes low along with shortening the mean life span. It is not preferable, on the other hand, that the  $\text{BaO}/\text{TiO}_2$  ratio  $m$  is over 1.035 as in the samples No. 2309 and 2310 since measurements becomes impossible due to insufficient sintering. Accordingly, the  $\text{BaO}/\text{TiO}_2$  ratio  $m$  in the range of  $1.000 < m \leq 1.035$  is preferable.

It is not preferable that the amount of addition of the first or second side component is zero as in the samples No. 2311 and 2313 because measurements are impossible due to insufficient sintering. It is not preferable that the amount of addition of the first or second side component exceeds 3.0 parts by weight as seen in the samples No. 2312 and 2314, on the other hand, because the dielectric loss exceeds 1.0% and the insulation resistance and insulation breakdown voltage are lowered along with shortening the mean life span. Accordingly, the preferable content of either the first or the second components is 0.2 to 3.0 parts by weight.

The contents of the alkali earth metal oxides contained in barium titanate as impurities are suppressed below 0.02% by weight because, when the contents of the alkali earth metal oxides exceeds 0.02% by weight as in the sample No. 2315, the dielectric constant is decreased.

#### (Example 17)

A material with a composition of  $\text{BaO}_{1.010} \cdot \text{TiO}_2 + 0.015\text{Ho}_2\text{O}_3 + 0.01\text{BaZrO}_3 + 0.03 \text{ MnO}$  (mole ratio) was prepared using barium titanate in TABLE 1A as a dielectric powder. A monolithic ceramic capacitor was produced by the same method as in Example 1, except that an oxide represented by  $\text{Li}_2\text{O}-(\text{Si}, \text{Ti})\text{O}_2\text{-MO}$  shown in Table 2006, having a mean particle size of 1  $\mu\text{m}$  or less produced by heating the material described above at 1200 to 1500 °C, was added as the first side component. The overall dimensions of the monolithic ceramic capacitor produced is the same as in Example 1. The electric characteristics were measured by the same method as in Example 1. The results are shown in TABLE 2014. In Table 2014, the samples No. 2401 to 2422 correspond to the samples No. 2101 to 2122 in TABLE 2006. For example, the sample No. 2401 in TABLE 2014 was obtained by adding the side component of the sample No. 2101 in TABLE 2006.

Table 2014

Sample No.	Baking temp. (°C)	Dielectric constant	Dielectric loss tan δ (%)	Ratio of temperature dependent capacitance change (%)					DC bias charac-teristic (%)	Product CR (Ω · F)				Insulation breakdown voltage (kV/mm)		Humidity resistance load test: Number of rejection	Mean life span (h)
				ΔC/C <sub>25</sub>						315V Im-pressed Voltage	945V Im-pressed Voltage	315V Im-pressed Voltage	945V Im-pressed Voltage	AC	DC		
				ΔC/C <sub>20</sub>		ΔC/C <sub>55</sub>		ΔC/C <sub>125</sub>									
				-25°C	85°C	-55°C	125°C				25°C	150°C					
2401	1300	1430	0.7	2.6	-8.4	3.6	-10.1	10.1	-36	5100	4850	220	210	12	14	0/72	880
2402	1300	1430	0.6	2.3	-8.4	3.4	-10.5	10.5	-36	5120	4860	210	200	12	14	0/72	900
2403	1280	1440	0.7	2.5	-8	4.1	-10.6	10.6	-36	5230	4970	200	190	12	14	0/72	870
2404	1300	1470	0.7	2.2	-8.1	4.2	-11	11	-38	5100	4850	230	220	12	14	0/72	820
2405	1300	1430	0.7	2.2	-7.9	4	-11.3	11.3	-36	5160	4900	230	220	13	15	0/72	820
2406	1280	1430	0.7	2.7	-8	4.2	-11.5	11.5	-36	5230	4970	210	200	12	14	0/72	950
2407	1300	1430	0.7	2.6	-8.2	3.9	-10.5	10.5	-36	5140	4880	220	210	13	15	0/72	830
2408	1280	1460	0.6	2.8	-8.3	4.1	-11	11	-37	5150	4890	220	210	12	14	0/72	800
2409	1280	1460	0.7	2.4	-8	4.2	-12.1	12.1	-37	5120	4860	200	190	12	14	0/72	880
2410	1280	1480	0.7	2.3	-8.1	4.1	-11.7	11.7	-38	5130	4870	220	210	12	14	0/72	850
2411	1300	1440	0.7	2.4	-8.2	3.9	-11.6	11.6	-36	5140	4880	220	210	12	14	0/72	810
2412	1280	1420	0.6	2.2	-8.3	3.8	-11.5	11.5	-36	5260	5000	200	190	12	14	0/72	920
2413	1350								Unmeasurable due to insufficient sintering								
2414	1350								Unmeasurable due to insufficient sintering								
2415	1350	1440	1.8	2.8	-8	3.8	-11	11	-38	5220	4960	190	180	11	12	55/72	120
2416	1350								Unmeasurable due to insufficient sintering								
2417	1350								Unmeasurable due to insufficient sintering								
2418	1300	1460	0.8	2.7	-8.2	4.1	-10.7	10.7	-37	5170	4910	210	200	11	13	0/72	860
2419	1350	1430	1.6	2.5	-8.3	4.2	-10.8	10.8	-36	5280	5020	220	210	11	13	33/72	110
2420	1300	1440	0.7	2.5	-8.4	4	-11.5	11.5	-37	5100	4850	190	180	12	13	0/72	870
2421	1350	1460	1.8	2.7	-8.3	3.9	-10	10	-37	5160	4900	190	180	11	13	52/72	130
2422	1350	1420	1.6	2.5	-8.2	3.7	-10.7	10.7	-36	5200	4940	200	190	11	13	49/72	140

As is evident from the samples No. 2401 to 2412, 2418 and 2420, preferable results are obtained in the samples in which oxides of the samples No. 2101 to 2112, 2118 and 2120 in TABLE 2006 with compositions within or on the bound-

any lines of the area surrounded by the straight lines connecting each spot indicated by A ( $x = 20, y = 80, z = 0$ ), B ( $x = 10, y = 80, z = 10$ ), C ( $x = 10, y = 70, z = 20$ ), D ( $x = 35, y = 45, z = 20$ ), E ( $x = 45, y = 45, z = 10$ ) and F ( $x = 45, y = 55, z = 0$ ) (wherein  $x, y$  and  $z$  represent mole %,  $w$  represents mole ratio,  $w$  being within the range of  $0.3 \leq w < 1.0$  when it falls on the line A - F) in the three component phase diagram of the oxides represented by  $\text{Li}_2\text{O}-(\text{Si}_w, \text{Ti}_{1-w})\text{O}_2\text{-MO}$  in FIG. 4 are added, wherein the samples have a capacitance decreasing ratio of as small as within -40% at an impressed voltage of 5 kV/mm and a dielectric loss of 1.0% or less, along with the rate of change of the electrostatic capacitance against temperature changes satisfying the B-level characteristic standard stipulated in the JIS Standard in the temperature range of -25 °C to +85 °C and X7R-level characteristic standard stipulated in the EIA standard in the temperature range of -55 °C to +125 °C.

Moreover, the insulation resistances at 25 °C and 150 °C as expressed by the product CR show as high values as 5000  $\Omega \cdot \text{F}$  or more and 190  $\Omega \cdot \text{F}$  or more, respectively, when the ceramic capacitor is used under a high electric field strength of 10 kV/mm. The insulation breakdown voltage also shows high values of 12 kV/mm or more under the AC voltage and 14 kV/mm or more under the DC voltage. In addition, an acceleration test at 150 °C and DC 25 kV/mm gave a mean life span as long as 800 hours or more besides enabling a relatively low firing temperature of 1300 °C or less.

When the oxide represented by  $\text{Li}_2\text{O}-(\text{Si}_w, \text{Ti}_{1-w})\text{O}_2\text{-MO}$  has a composition outside of the composition described above as in the samples No. 2113 to 2117 and 2119, on the other hand, sintering becomes insufficient or many rejection appear in the humidity resistance load test even after sintering as seen in the samples No. 2413 to 2417 and 2419 in TABLE 2014. When the composition falls on the line A - F and  $w = 1.0$  as in the samples No. 2119 and 2121 in TABLE 2006, the sintering temperature becomes high and many rejects appear in the humidity resistance load test. When the value of  $w$  is less than 0.30 as in the sample No. 2122 in TABLE 2006, the sintering temperature becomes high and many rejects appear in the humidity resistance load test as seen in the sample No. 2422 in TABLE 2014.

(Example 18)

A material with a composition of  $\text{BaO}_{1.010} \cdot \text{TiO}_2 + 0.025\text{Eu}_2\text{O}_3 + 0.01\text{BaZrO}_3 + 0.05 \text{ MnO}$  (mole ratio) was prepared using barium titanate "A" in TABLE 1 as a dielectric powder. A monolithic ceramic capacitor was produced by the same method as in Example 1, except that an oxide  $\text{Si}_2\text{O-TiO}_2\text{-XO}$  shown in Table 2008, having a mean particle size of 1  $\mu\text{m}$  or less produced by heating the material described above at 1200 to 1500 °C, was added as the second side component. The overall dimensions of the monolithic ceramic capacitor produced is the same as in Example 1. The electric characteristics were measured by the same method as in Example 1. The results are shown in TABLE 2015. In Table 2015, the samples No. 2501 to 2519 in TABLE 2015 correspond to the samples No. 2201 to 2219 in TABLE 2008. For example, the sample No. 2501 in TABLE 2015 was obtained by adding the side component of the sample No. 2201 in TABLE 2008.

Table 2015

Sample No.	Baking temp. (°C)	Dielectric constant	Dielectric loss tan δ (%)	Ratio of temperature dependent capacitance change (%)					DC bias characteristic (%) ΔC/C 5kV/mm	Product CR (Ω · F)				Insulation breakdown voltage (kV/mm)		Humidity resistance load test: Number of reject	Mean life span (h)
				ΔC/C <sub>20</sub>		ΔC/C <sub>25</sub>		Maximum value		315V Im <sup>2</sup> pressed Voltage	945V Im <sup>2</sup> pressed Voltage	315V Im <sup>2</sup> pressed Voltage	945V Im <sup>2</sup> pressed Voltage				
				-25°C	85°C	-55°C	125°C										
															25°C		
2501	1300	1350	0.9	2.2	-8.3	3.6	-10.2	10.2	-32	5120	4860	230	220	12	14	0/72	870
2502	1300	1350	0.8	2.3	-8.6	4.1	-10.4	10.4	-32	5210	4950	240	230	12	14	0/72	920
2503	1300	1360	0.9	2.4	-8.7	4	-11	11	-32	5130	4870	220	210	12	14	0/72	860
2504	1300	1320	0.8	2.3	-9	3.8	-11.6	11.6	-31	5140	4880	230	220	13	15	0/72	840
2505	1300	1360	0.9	2.2	-8.2	3.9	-10.8	10.8	-32	5160	4900	220	210	12	14	0/72	920
2506	1300	1340	0.8	2.3	-8.6	4	-10.7	10.7	-32	5120	4860	210	200	12	14	0/72	910
2507	1300	1310	0.9	2.6	-8.4	4.2	-11	11	-30	5240	4980	230	220	13	15	0/72	880
2508	1300	1340	0.9	2.5	-8.2	4	-11.5	11.5	-32	5230	4970	220	210	12	14	0/72	900
2509	1300	1330	0.9	2.3	-8.3	3.7	-11.6	11.6	-31	5200	4940	220	210	12	14	0/72	920
2510	1300	1370	0.9	2.1	-8.4	3.5	-11.3	11.3	-33	5030	4780	210	200	12	14	0/72	930
2511	1300	1360	0.8	2.2	-8.6	3.8	-10.9	10.9	-32	5430	5160	340	320	12	14	0/72	850
2512	1300	1350	0.9	2.6	-8.3	3.6	-10.7	10.7	-32	5410	5140	330	310	12	14	0/72	900
2513	1350	1330	0.9	2.4	-8.4	4	-11	11	-31	5120	4860	220	210	11	13	55/72	120
2514	1350			Unmeasurable due to insufficient sintering													
2515	1350			Unmeasurable due to insufficient sintering													
2516	1350	1320	1	2.3	-8.5	3.8	-11.5	11.5	-30	5160	4900	230	220	11	13	64/72	110
2517	1350			Unmeasurable due to insufficient sintering													
2518	1350			Unmeasurable due to insufficient sintering													
2519	1350			Unmeasurable due to insufficient sintering													

As is evident from the samples No. 2501 to 2512 in TABLE 2015, preferable results are obtained in the samples in which oxides of the samples No. 2201 to 2212 in TABLE 2008 with compositions within or on the boundary lines of the area surrounded by the straight lines connecting each spot indicated by A ( $x = 85$ ,  $y = 1$ ,  $z = 14$ ), B ( $X = 35$ ,  $y = 51$ ,  $z =$

14), C (X = 30, y = 20, z = 50) and D (X = 39, y = 1, z = 60) (wherein x, y and z represent mole %) in the three component phase diagram of the  $\text{SiO}_2\text{-TiO}_2\text{-XO}$  oxides shown in FIG. 5 are added, wherein the samples have a capacitance decreasing ratio of as small as within -40% at an impressed voltage of 5 kV/mm and a dielectric loss of 1.0% or less, along with the rate of change of the electrostatic capacitance against temperature changes satisfying the B-level characteristic standard stipulated in the JIS Standard in the temperature range of -25 °C to +85 °C and X7R-level characteristic standard stipulated in the EIA standard in the temperature range of -55 °C to +125 °C.

Moreover, the insulation resistances at 25 °C and 150 °C as expressed by the product CR show as high values as 5000  $\Omega \cdot \text{F}$  or more and 200  $\Omega \cdot \text{F}$  or more, respectively, when the ceramic capacitor is used under a high electric field strength of 10 kV/mm. The insulation breakdown voltage also shows high values of 12 kV/mm or more under the AC voltage and 14 kV/mm or more under the DC voltage. In addition, an acceleration test at 150 °C and DC 25 kV/mm gave a mean life span as long as 800 hours or more besides enabling a relatively low firing temperature of 1300 °C or less.

When the oxide  $\text{SiO}_2\text{-TiO}_2\text{-XO}$  has a composition outside of the composition described above as in the samples No. 2213 to 2119 in TABLE 2008, on the other hand, sintering becomes insufficient or many rejection appear in the humidity resistance load test even after sintering as seen in the samples No. 2513 to 2519 in TABLE 2015.

While a monolithic capacitor having an insulation resistance of 5400  $\Omega \cdot \text{F}$  or more and 330  $\Omega \cdot \text{F}$  or more at 25 °C and 150 °C, respectively, under a strong electric field of 10 kV/mm can be obtained by allowing  $\text{Al}_2\text{O}_3$  and/or  $\text{ZrO}_2$  to contain in the  $\text{SiO}_2\text{-TiO}_2\text{-XO}$  oxides as in the sample No. 2211 and 2212 in TABLE 2008, sintering property is extremely decreased as shown in the samples NO. 2517 and 2518 in TABLE 2015 when  $\text{Al}_2\text{O}_3$  and  $\text{ZrO}_2$  are added in an amounts of 15 parts by weight or more and 5 parts by weight or more, respectively, as in the samples No. 2517 and 2518 in TABLE 2015.

Although powders prepared by the oxalic acid method are used in the foregoing examples, the methods are not limited thereto but a powder of barium titanate prepared by an alkoxide method or hydrothermal synthesis method may be used. It may happen that the characteristics of the monolithic ceramic capacitor are more improved than those shown in the foregoing examples by using these powders.

The oxide powders as starting materials are not limited to those hitherto described, but the resulting characteristics are not affected in any sense by using a solution of an alkoxide or organometallic compound, provided that the starting materials are formulated so as to construct the dielectric ceramic layers within the scope of the present invention.

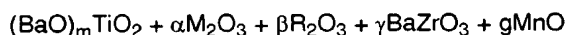
It can be made clear from the foregoing descriptions that the dielectric ceramic composition according to the present invention is not reduced by firing in the reducing atmosphere along with not being formed into semiconductors, besides sintering is possible at a relatively low temperature of 1300 °C or less.

Accordingly, when a monolithic ceramic capacitor is constructed by using this dielectric ceramic composition as dielectric ceramic layers, the production cost of the monolithic ceramic capacitor can be reduced since base metals such as nickel or nickel alloys may be used for the electrode materials.

When the monolithic ceramic capacitor using this dielectric ceramic composition is used under a high electric field of 10 kV/mm where reliability can not be ensured due to low insulation resistance in the monolithic ceramic capacitor using nickel or nickel alloys for the conventional inner electrodes, a monolithic ceramic capacitor being excellent in weather resistance properties such as high temperature load at an impressed voltage of DC 25 kV/mm at 150 °C and humidity resistance load can be obtained, wherein the insulation resistances at room temperature and at 150 °C represented by a product of the insulation resistance and electrostatic capacitance (a CR product) becomes as high as 4900 to 5000  $\Omega \cdot \text{F}$  and 190 to 200  $\Omega \cdot \text{F}$ , respectively, the voltage dependency of the insulation resistance is low, the capacitance decrease ratio at an impressed voltage of 5 kV/mm is as small as 40 to 45% and the insulation durability is high, besides the temperature characteristics of the electrostatic capacitance satisfies the B-level characteristic standard stipulated in the JIS Standard and X7R-level characteristic standard stipulated in the EIA standard.

## Claims

1. A dielectric ceramic composition comprising barium titanate containing 0.02% by weight or less of alkali metal oxides, at least one of either scandium oxide or yttrium oxide, at least one kind of compound selected from europium oxide, gadolinium oxide, terbium oxide and dysprosium oxide, barium zirconate and manganese oxide, containing an essential component represented by the following composition formula;

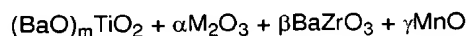


(wherein  $\text{M}_2\text{O}_3$  represents at least one of either  $\text{Sc}_2\text{O}_3$  or  $\text{Y}_2\text{O}_3$  and  $\text{R}_2\text{O}_3$  represents at least one of the compound selected from  $\text{Eu}_2\text{O}_3$ ,  $\text{Gd}_2\text{O}_3$ ,  $\text{Tb}_2\text{O}_3$  and  $\text{Dy}_2\text{O}_3$ ,  $\alpha$ ,  $\beta$ ,  $\gamma$  and  $g$  representing mole ratio in the range of  $0.001 \leq \alpha \leq 0.05$ ,  $0.001 \leq \beta \leq 0.05$ ,  $0.005 \leq \gamma \leq 0.06$ ,  $0.001 < g \leq 0.13$  and  $\alpha + \beta \leq 0.06$  with  $1.000 < m \leq 1.035$ ),

characterized by containing 0.2 to 3.0 parts by weight of either the first or second side component relative to 100 parts by weight of said essential component, wherein said first side component is an oxide represented by

$\text{Li}_2\text{O} - (\text{Si}, \text{Ti})\text{O}_2 - \text{MO}$  (wherein MO is at least one of  $\text{Al}_2\text{O}_3$  or  $\text{ZrO}_2$ ) and said second side component is an oxide represented by  $\text{SiO}_2 - \text{TiO}_2 - \text{XO}$  (wherein XO is at least one of the compound selected from BaO, CaO, SrO, MgO, ZnO and MnO).

2. A dielectric ceramic composition according to Claim 1, wherein said essential component further contains h mole ratio of magnesium oxide, where  $0.001 < g \leq 0.12$ ,  $0.001 < h \leq 0.12$  and  $g + h \leq 0.13$
3. A dielectric ceramic composition comprising barium titanate containing 0.02% by weight or less of alkali metal oxides, at least one of either scandium oxide or yttrium oxide, barium zirconate and manganese oxide, containing an essential component represented by the following composition formula;



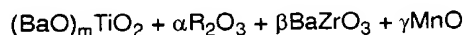
(wherein  $\text{M}_2\text{O}_3$  represents at least one of either  $\text{Sc}_2\text{O}_3$  or  $\text{Y}_2\text{O}_3$ ,  $\alpha$ ,  $\beta$  and  $\gamma$  representing mole ratio in the range of  $0.001 \leq \alpha \leq 0.06$ ,  $0.005 \leq \beta \leq 0.06$  and  $0.001 < \gamma \leq 0.13$  with  $1.000 < m \leq 1.035$ ),

characterized by containing 0.2 to 3.0 parts by weight of either the first or second side component relative to 100 parts by weight of said essential component, wherein said first side component is an oxide represented by  $\text{Li}_2\text{O} - (\text{Si}, \text{Ti})\text{O}_2 - \text{MO}$  (wherein MO is at least one of  $\text{Al}_2\text{O}_3$  or  $\text{ZrO}_2$ ) and said second side component is an oxide represented by  $\text{SiO}_2 - \text{TiO}_2 - \text{XO}$  (wherein XO is at least one of the compound selected from BaO, CaO, SrO, MgO, ZnO and MnO).

4. A dielectric ceramic composition according to Claim 3, wherein said essential component further contains g mole ratio of magnesium oxide, where  $0.001 < \gamma \leq 0.12$ ,  $0.001 < g \leq 0.12$  and  $\gamma + g \leq 0.13$

5. A dielectric ceramic composition comprising:

barium titanate containing 0.02% by weight or less of alkali metal oxides, at least one kind of the compound selected from europium oxide, gadolinium oxide, terbium oxide, dysprosium oxide, holmium oxide, erbium oxide, thulium oxide and ytterbium oxide, barium zirconate and manganese oxide, containing an essential component represented by the following composition formula;



(wherein  $\text{R}_2\text{O}_3$  represents at least one kind of compound selected from  $\text{Eu}_2\text{O}_3$ ,  $\text{Gd}_2\text{O}_3$ ,  $\text{Tb}_2\text{O}_3$ ,  $\text{Dy}_2\text{O}_3$ ,  $\text{Ho}_2\text{O}_3$ ,  $\text{Er}_2\text{O}_3$ ,  $\text{Tm}_2\text{O}_3$  and  $\text{Yb}_2\text{O}_3$ ,  $\alpha$ ,  $\beta$  and  $\gamma$  representing mole ratio in the range of  $0.001 \leq \alpha \leq 0.06$ ,  $0.005 \leq \beta \leq 0.06$  and  $0.001 < \gamma \leq 0.13$  with  $1.000 < m \leq 1.025$ ),

characterized by containing 0.2 to 3.0 parts by weight of either the first or second side component relative to 100 parts by weight of said essential component, wherein said first side component is an oxide represented by  $\text{Li}_2\text{O} - (\text{Si}, \text{Ti})\text{O}_2 - \text{MO}$  (wherein MO is at least one of  $\text{Al}_2\text{O}_3$  or  $\text{ZrO}_2$ ) and said second side component is an oxide represented by  $\text{SiO}_2 - \text{TiO}_2 - \text{XO}$  (wherein XO is at least one of the compound selected from BaO, GaO, SrO, MgO, ZnO and MnO).

6. A dielectric ceramic composition according to Claim 5, wherein said essential component further contains g mole ratio of magnesium oxide, where  $0.001 \leq \beta \leq 0.06$ ,  $0.001 < \gamma \leq 0.12$ ,  $0.001 < g \leq 0.12$  and  $\gamma + g \leq 0.13$ .

7. A dielectric ceramic composition according to Claim 1 to Claim 6, wherein said first side component, when its composition is represented by  $x\text{Li}_2\text{O} - y(\text{Si}_w\text{Ti}_{1-w})\text{O}_2 - z\text{MO}$  (wherein x, y and z represent mol% and w is in the range of  $0.30 \leq w \leq 1.00$ ), falls within or on the boundary lines of the area surrounded by the straight lines connecting each point indicated by A ( $x = 20$ ,  $y = 80$ ,  $z = 0$ ), B ( $x = 10$ ,  $y = 80$ ,  $z = 10$ ), C ( $x = 10$ ,  $y = 70$ ,  $z = 20$ ), D ( $x = 35$ ,  $y = 45$ ,  $z = 20$ ), E ( $x = 45$ ,  $y = 45$ ,  $z = 10$ ) and F ( $x = 45$ ,  $y = 55$ ,  $z = 0$ ) (when the composition falls on the straight line of A - F, w is within the area of  $0.3 \leq w < 1.0$ ) in the three component phase diagram defined by the apexes corresponding to each component.

8. A dielectric ceramic composition according to Claim 1 to Claim 6, wherein said second side component, when its composition is represented by  $x\text{SiO}_2 - y\text{TiO}_2 - z\text{XO}$  (wherein x, y and z represent mol%), falls within or on the boundary lines of the area surrounded by straight lines connecting each point indicated by A ( $x = 85$ ,  $y = 1$ ,  $z = 14$ ), B ( $x = 35$ ,  $y = 51$ ,  $z = 14$ ), C ( $x = 30$ ,  $y = 20$ ,  $z = 50$ ) and D ( $x = 39$ ,  $y = 1$ ,  $z = 60$ ) in the three component phase diagram defined by the apexes corresponding to each component.

9. A dielectric ceramic composition according to Claim 8, wherein the second side component contains in total of 15 parts by weight or less of at least one of  $\text{Al}_2\text{O}_3$  and  $\text{ZrO}_2$  (the content of  $\text{ZrO}_2$  is 5 parts by weight or less) relative to 100 parts by weight of the oxide represented by  $\text{SiO}_2\text{-TiO}_2\text{-XO}$ .
- 5 10. A monolithic ceramic capacitor provided with a plurality of dielectric ceramic layers, inner electrodes formed between said ceramic layers and outer electrodes being electrically connected to said inner electrodes, wherein said dielectric ceramic layers are constructed by the dielectric ceramic composition according to any one of Claim 1 to Claim 6 and said inner electrodes are constructed by nickel or a nickel alloy.
- 10 11. A monolithic ceramic capacitor according to Claim 10, wherein the outer electrode is provided with a sintered layer of an electroconductive metal powder or an electroconductive metal powder supplemented with glass frits.
12. A monolithic ceramic capacitor according to Claim 10, wherein the outer electrode is provided with a first layer comprising a sintered layer of the electroconductive metal powder or the electroconductive metal powder supplemented  
15 with glass frits and a second layer comprising a plating layer thereon.
13. A monolithic ceramic capacitor according to Claim 11, wherein the outer electrode is provided with a first layer comprising a sintered layer of the electroconductive metal powder or the electroconductive metal powder supplemented  
20 with glass frits and a second layer comprising a plating layer thereon.

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Fig. 1

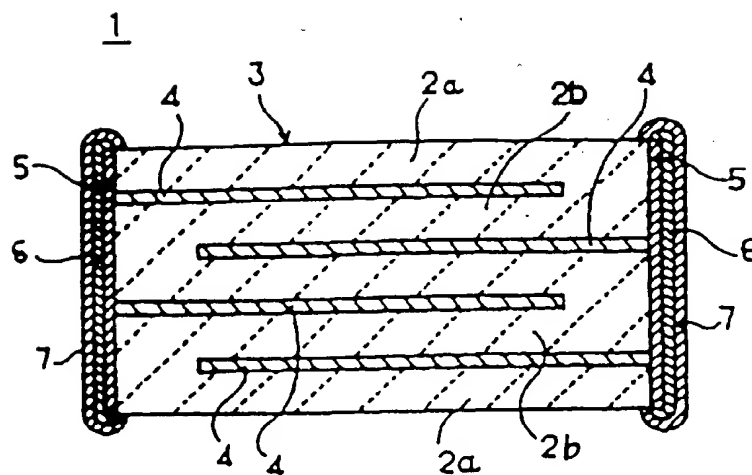


Fig. 2

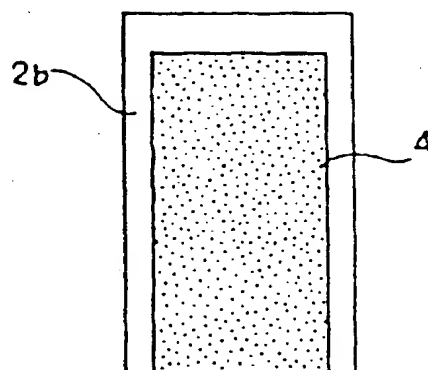


Fig. 3

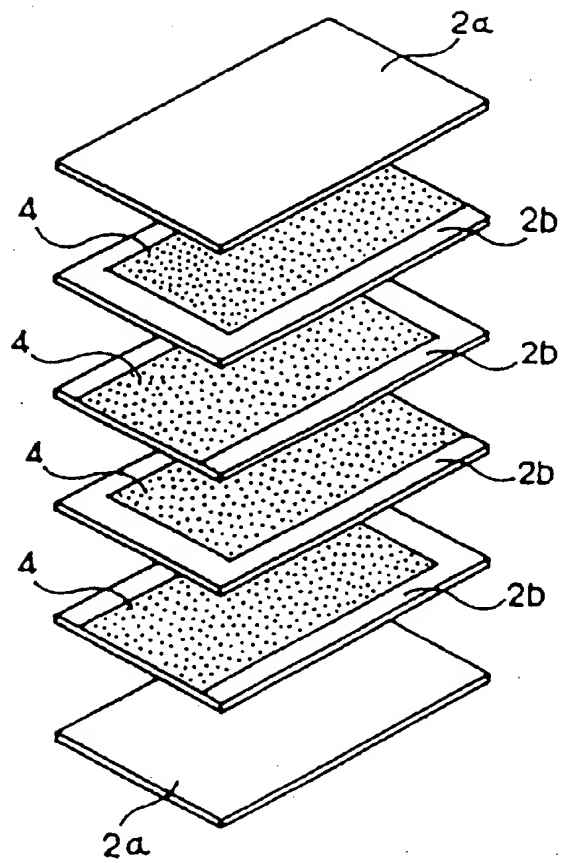


Fig. 4

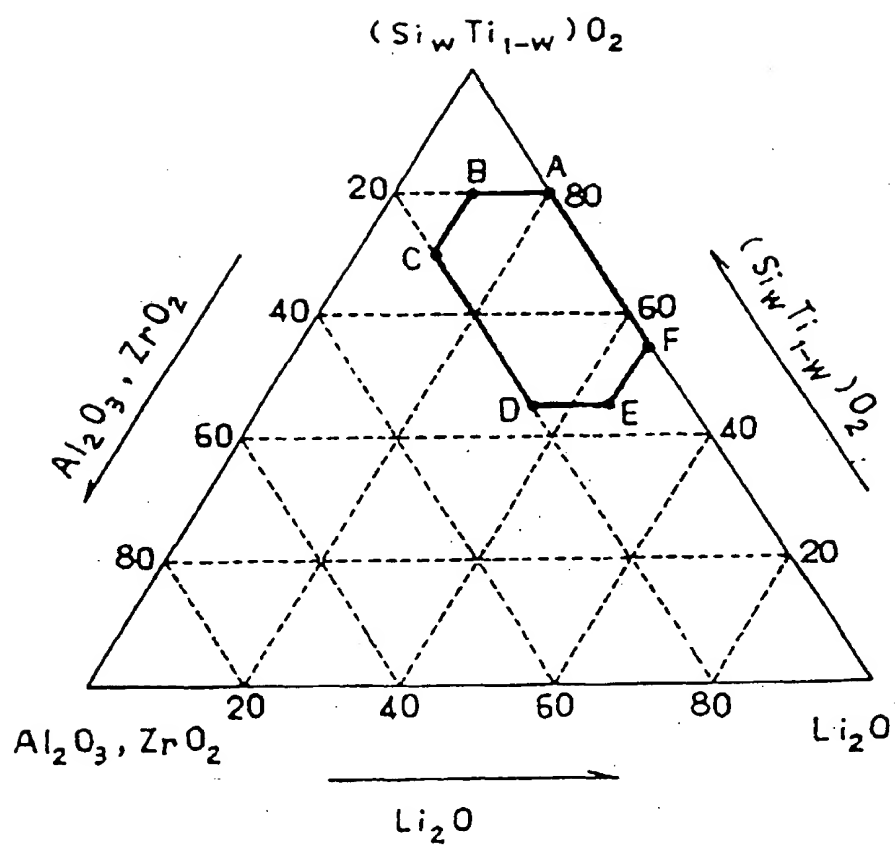
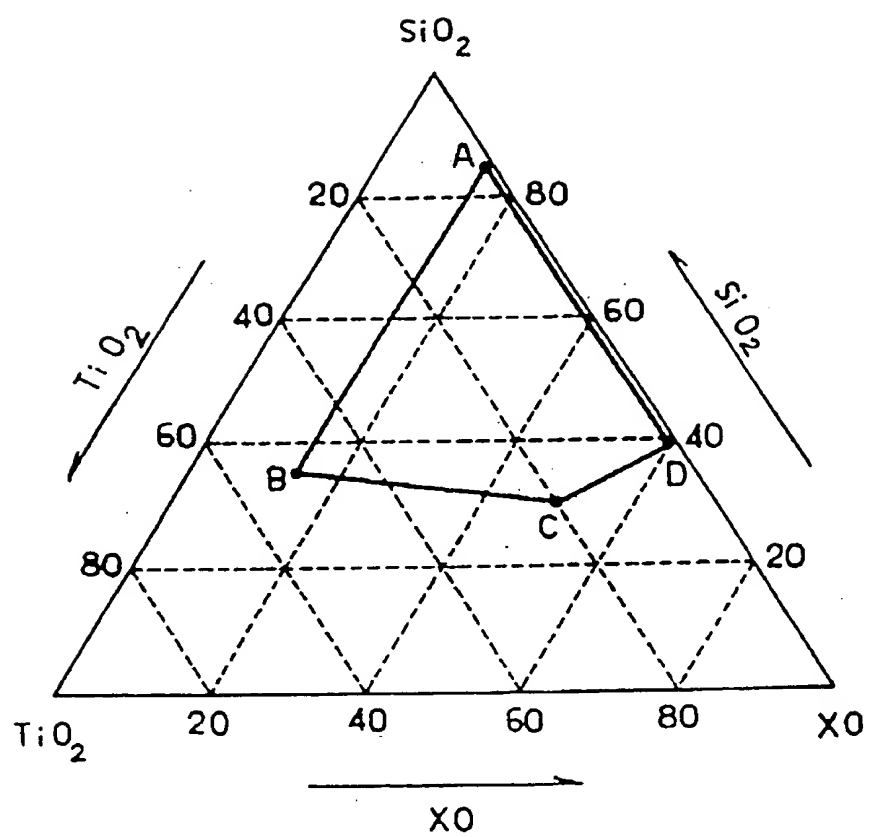


Fig. 5





European Patent  
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# EUROPEAN SEARCH REPORT

Application Number  
EP 98 11 3795

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
X	EP 0 737 655 A (MURATA MANUFACTURING CO., LTD.) 16 October 1996 * page 2, line 58 - page 4, line 50 * * claims 1-20; example 2; tables 7-10 * ---	5-7, 10-13	C04B35/468 H01G4/12 H01B3/12
A	EP 0 605 904 A (MURATA MANUFACTURING CO., LTD.) 13 July 1994 * page 3, line 2 - line 18; claim 14; example 1; table 1 * -----	1-13	
			TECHNICAL FIELDS SEARCHED (Int.Cl.6)
			C04B H01G H01B
The present search report has been drawn up for all claims			
Place of search <b>THE HAGUE</b>		Date of completion of the search <b>27 October 1998</b>	Examiner <b>Luethe, H</b>
<p>CATEGORY OF CITED DOCUMENTS</p> <p>X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document</p> <p>T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons S : member of the same patent family, corresponding document</p>			

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